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CHARACTERISTICS OF FERNALD'S K-65 RESIDUE BEFORE, DURING AND AFTER VITRIFICATION FEBRUARY 1991

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REPORT

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K-65 RESIDUE BEFORE, DURING AND AFTER
VITRIFICATION

by

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Battelle Pacific Northwest Labs

Richland, Washington

February 1991

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SUMMARY

Motivation for Vitrification

Vitrification of radioactive and hazardous wastes has been under thorough investigation since the mid-1950s. During the high-level waste development program, the U.S. Department of Energy accumulated over 40 years of operating experience with the vitrification process (Chapman and McElroy, 1989). Vitrification has endured international scrutiny and is the preferred international treatment method for the most radioactive and hazardous high-level radioactive wastes (DOE/RL-90-27). Other compelling factors support the use of vitrification for treating many types of hazardous and radioactive wastes:

- The U.S. Environmental Protection Agency (EPA) has promulgated vitrification as the treatment standard {i.e., best demonstrated available technology (BDAT)} for high-level radioactive mixed waste (Federal Register, June 1, 1991), and a BDAT for arsenic-containing hazardous wastes (Federal Register, ca. May, 1990).

- The glass, formed with, at most, minor chemical additions to the waste, generally tests by the Toxicity Characteristic Leachate Procedure (TCLP) or by the Extraction Procedure (EP) toxicity criteria as nonhazardous.

- Volume reduction for solids is typically greater than 60%.

For these fundamental reasons, DOE has constructed waste vitrification facilities at both Savannah River and West Valley, and is finalizing the design for a plant at Hanford. These fully remote plants will be operational in 1992, 1994, and 1999, respectively.

Vitrification is presently being investigated as a potential method for treating material from the Feed Materials Production Center in Fernald, Ohio. Two silos, designated as K-65, contain residues from the processing of pitchblende ores. This residue contains uranium, uranium daughter products, and some heavy metals (primarily lead).

Several factors will contribute to reducing the cost of vitrifying the K-65 residue. The K-65 residue is a simple oxide waste, very similar to conventional glass-making materials, and consequently does not require extensive glass formulation or glass former additions to produce a waste form.

With the simple addition of a reducing agent (e.g., graphite), 80% to 90% of the gold present in the K-65 residue (see Table 3) should be recoverable, and could offset the costs of treating the K-65 residue. Recovery of palladium, rhodium and ruthenium from simulated high-level wastes has been successfully demonstrated in both the laboratory and a small-scale melter by using reduced lead as the molten scavenging agent (Jensen, et. al., 1984). Because gold is less reactive with oxygen than any of the

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other elements mentioned previously, its recovery should be more readily achieved. In addition, recent innovations in melter construction and operation can significantly reduce the capital and operating costs over conventional industrial glass melters with similar production rates.

In a vitrified matrix, the diffusion of gases with atomic radii equal to or greater than krypton (1.03 angstrom) and xenon (1.24 angstrom), such as radon (1.34 angstrom), is nil (Sing and Swallow, 1960). Thus, once vitrified, release of radon from the residue will be limited to the modest amount of externally exposed surface area. Barretto (Barretto, 1975) found that volcanic glass has the highest radon retention ability of the 59 rock samples he studied. Based upon these favorable processing and product characteristics, vitrification of the K-65 residue is an environmentally progressive and technically sound option for treating this material.

Test Results

For the work reported here, Pacific Northwest Laboratory received approximately 7 kilograms of the K-65 residue from Silo 1 for vitrification tests. The objectives of the tests were to determine the quantity and composition of off-gas evolved during vitrification, the radon emanation rate from both the original K-65 residue and the vitrified product, and the leachability of the vitrified material.

- Vitrified K-65 residue (Specific Gravity = 3.1) has a volume that is 35% of dried, tamped K-65 residue (Specific Gravity = 1.06), a 65% volume reduction.

- The radon emanation flux from the K-65 residue was reduced by more than 33,000 times when vitrified. The flux from the original material was measured to be 1.5 million pCi/hr or 52,400 pCi/m²-s, while glass was 48 pCi/hr or 1.56 pCi/m²-s (an order of magnitude below the EPA limit of 20 pCi/m²-s). We predict that during full-scale processing, the flux may be further reduced by a total factor of up to 90,000 to 2,400,000 because the test crucible had both unmelted material and a coat of glass on the crucible walls. Therefore, the actual surface area exceeded the assumed surface area by a factor of more than 3. Furthermore, the tested glass sample had an exposed surface area-to-volume ratio that is 30 times higher than a filled 55-gallon drum, which is a likely casting form for actual processing conditions.

- The chemical composition of the off-gas released during processing is essentially that of air (see Table 6).

- The off-gas data indicate that for the chemicals present, 99.5% to 99.95% is retained in the glass. This is typical of results obtained during thousands of hours of melter testing with simulated high-level radioactive waste slurries.

- As measured by the TCLP, the vitrified K-65 residue tests as nonhazardous. The two TCLP heavy metals present in the glass were barium at 4.4 wt% and lead at 9.9 wt%. The leachate concentrations were 0.98 ppm and 0.3 ppm for barium and lead, respectively, which is well below the limits of 100 and 5 ppm for barium and lead. Results from EP toxicity tests for this K-65 residue show a leachate concentration of 0.76 and 630 ppm for barium and lead, respectively. Thus, the vitrified product improved the leach resistance for lead by a factor of over 2,000.

- The vitrified product is so durable that it could not be dissolved in a hot mixture of concentrated nitric and hydrofluoric acid by Controls for Environmental Pollution (CEP), Inc., during their analyses of the glass.

- The radon released during processing is only 23% of the total expected to be present at secular equilibrium, and could be easily absorbed in the off-gas treatment system by an activated charcoal bed (Fusamura, et al., 1963).

- Radon was measured to have an affinity for oils. This phenomenon may potentially be employed in a radon filtration/absorption scheme. For Dow Corning 704 Diffusion Pump Fluid and a contact time of about 0.006 seconds, over 50% of the radon was removed from the air stream. For Consolidated Vacuum Corporation's Convoil-20 Vacuum Pump Fluid with the same contact time, over 40% of the radon was removed. By providing greater contact time, very effective filtration/absorption may be achieved.

- The net emanation rate of radon was measured to decrease with higher concentrations of radon above the emanating source. This behavior has also been reported by Freeman (Freeman, 1981) and by Nielson (Nielson, et al., 1981). Although this information is outside the scope for this work, it does affect the way radon concentrations are monitored and interpreted.

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INTRODUCTION

Pacific Northwest Laboratory (PNL)[1] recently completed vitrification tests of material from the Feed Materials Production Center in Fernald, Ohio. Vitrification is the process of converting material to a glass form, and is attractive for application to wastes for many reasons: it produces an extremely durable, chemically stable glass product; it can greatly reduce the volume of waste; and it is a cost-effective treatment process. The vitrification process as it applies to treating wastes has been under development for years, and has recently been selected as the treatment standard for high-level radioactive mixed waste (Federal Register, June 1, 1990). It has also been recognized as a highly efficient method for treating wastes that contain arsenic (Federal Register, ca. May, 1990).

Two silos, designated as K-65, at the Fernald site contain residues from the processing of pitchblende ores. This residue contains uranium, uranium daughter products, and some heavy metals (primarily lead). PNL received approximately 7 kilograms of the K-65 residue for vitrification tests. The objectives of the tests were to determine the quantity and composition of off-gas evolved during vitrification, the radon emanation rate from both the original K-65 residue and the vitrified product, and the leachability of the vitrified material. To meet these objectives, samples of the residue were vitrified on a bench scale. The off-gas from the vitrification process was collected, measured (volume), and analyzed for radon and other constituents. The radon emanation rate from both the vitrified and nonvitrified residue was also measured.

This work was conducted in four phases. The following paragraphs describe these phases, and Figures 1 through 4 show the systems used for the work conducted in each phase. Figure 5 presents a flow diagram of the experimental procedure.

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[1] Pacific Northwest Laboratory is operated by Battelle Memorial Institute for the U.S. Department of Energy under Contract DE-AC06-76 RLO 1830.

Figure 1. OFF – GAS COLLECTION SYSTEM

GAS SAMPLE BOMBS

0.075 L
(GS)

RECIRCULATION LOOP

Pump

FLOW METER

INSTRUMENT AIR

TO HOOD

ACTIVATED CARBON COLUMN

Desiccant

COLLECTION SYSTEM

P = Pressure
T = Temperature
⊗ = Open Valve
● = Closed Valve

DISCHARGE LINE

Height Adjustment

SLURRIED K65 RESIDUE AND MELTING CRUCIBLE

2.5 L

FURNACE

COLLECTION VESSEL

CARBOY

GRADUATED CYLINDER (CS)
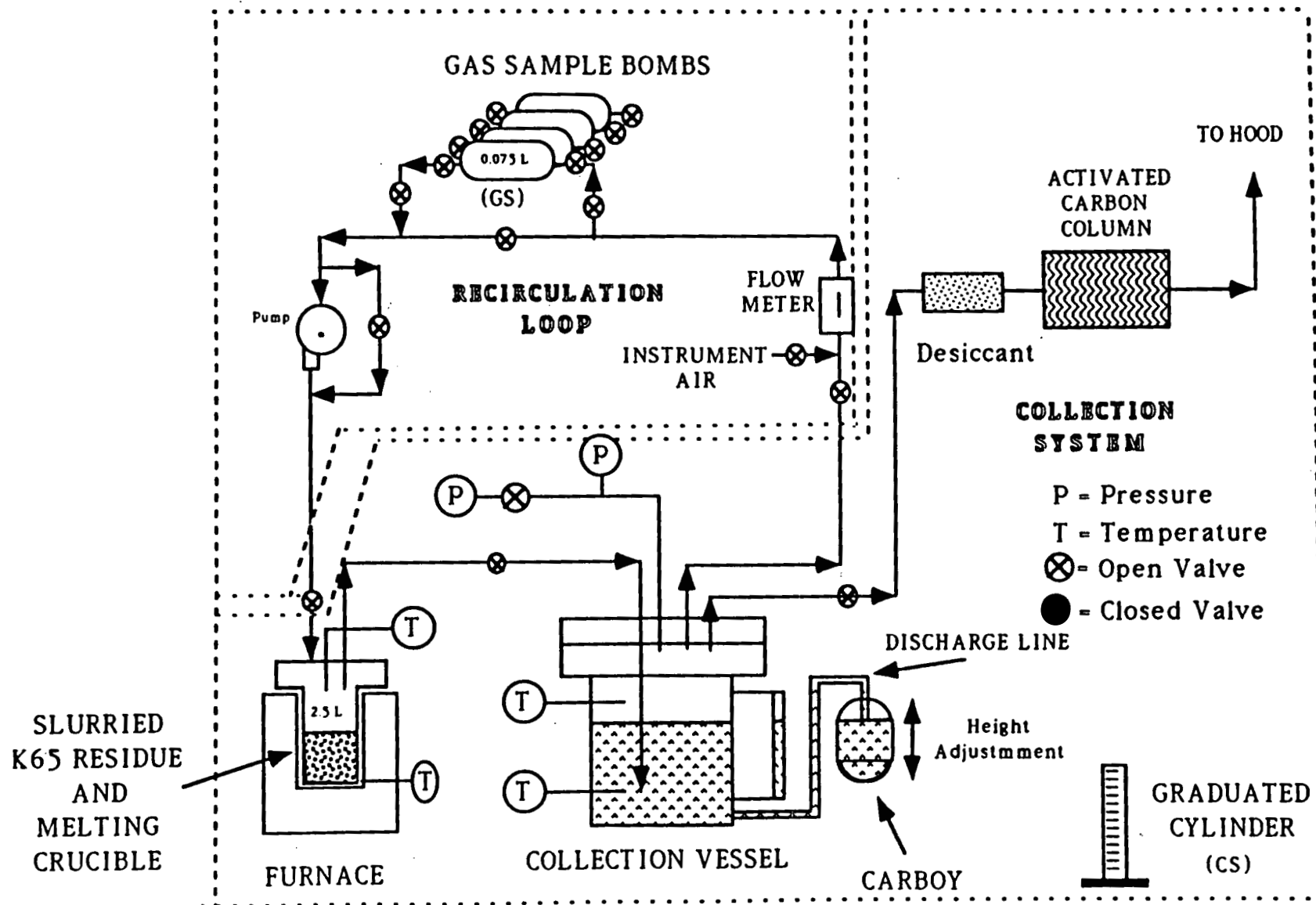
Figure 1a. SYSTEM FOR OFF-GAS COLLECTION DURING VITRIFICATION
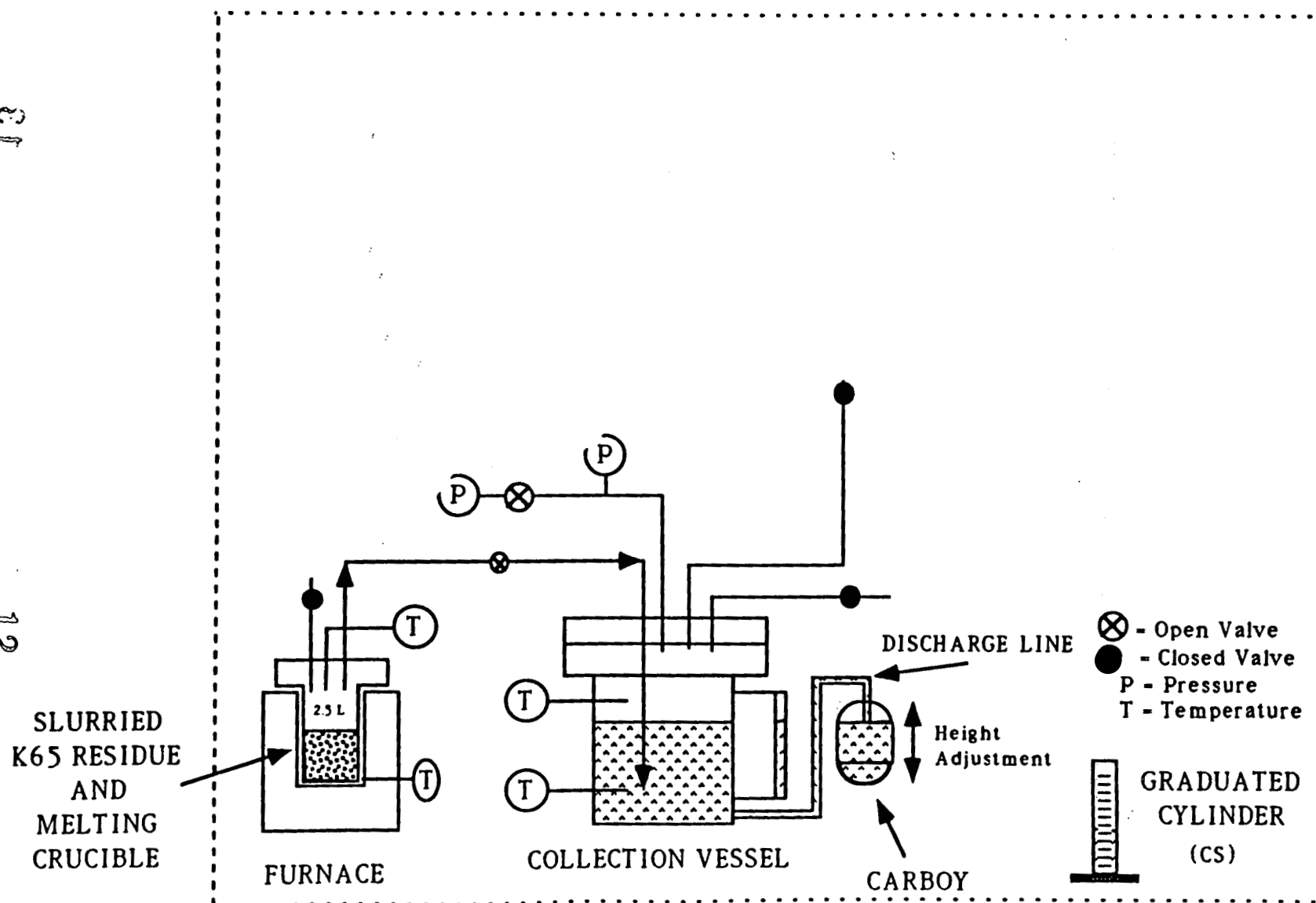
Figure 2. COLLECTED OFF - GAS SAMPLING ARRANGEMENT

Figure 2a. ON-SITE RADON MONITORING SYSTEM
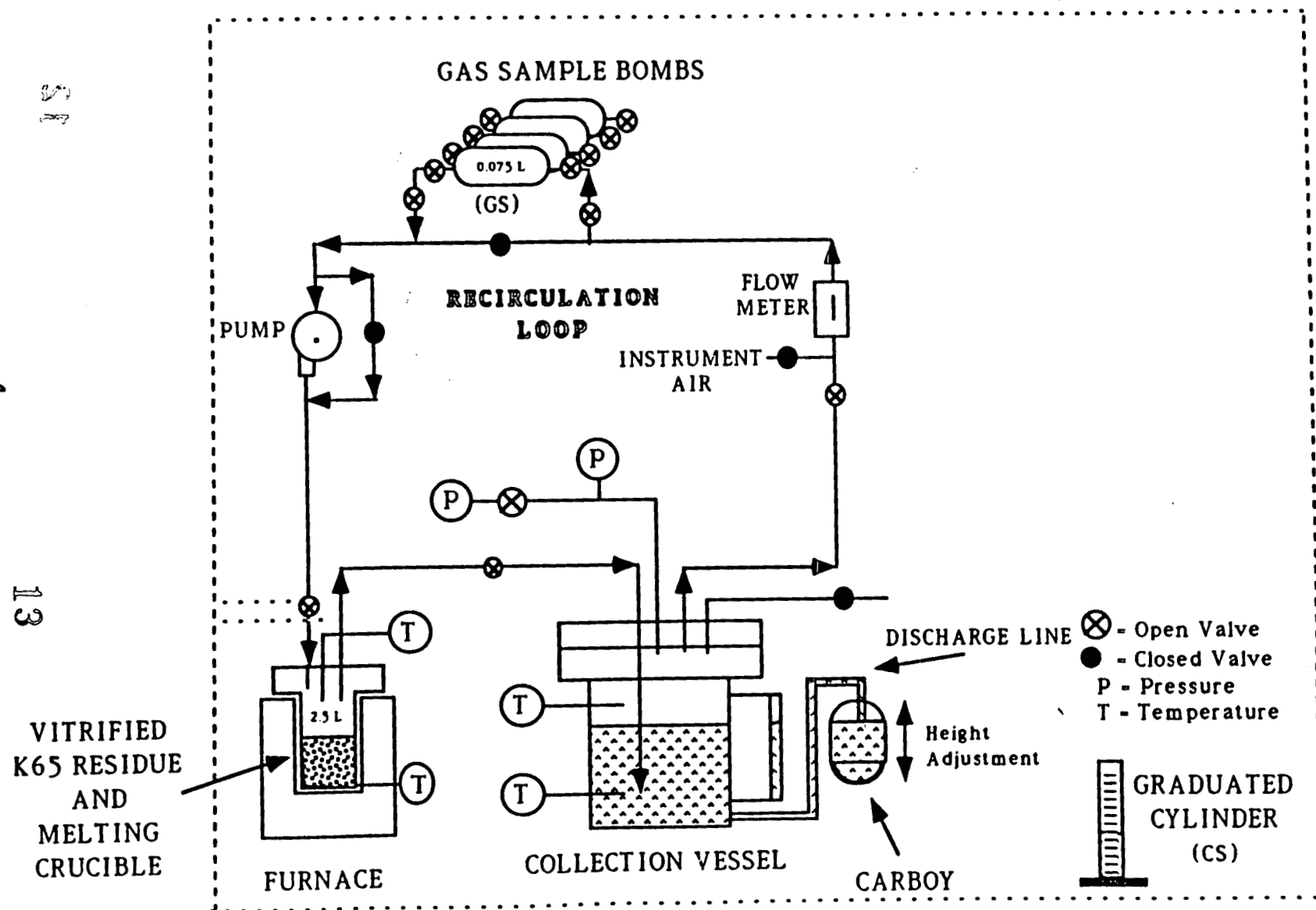
Figure 2a. ON-SITE RADON MONITORING SYSTEM
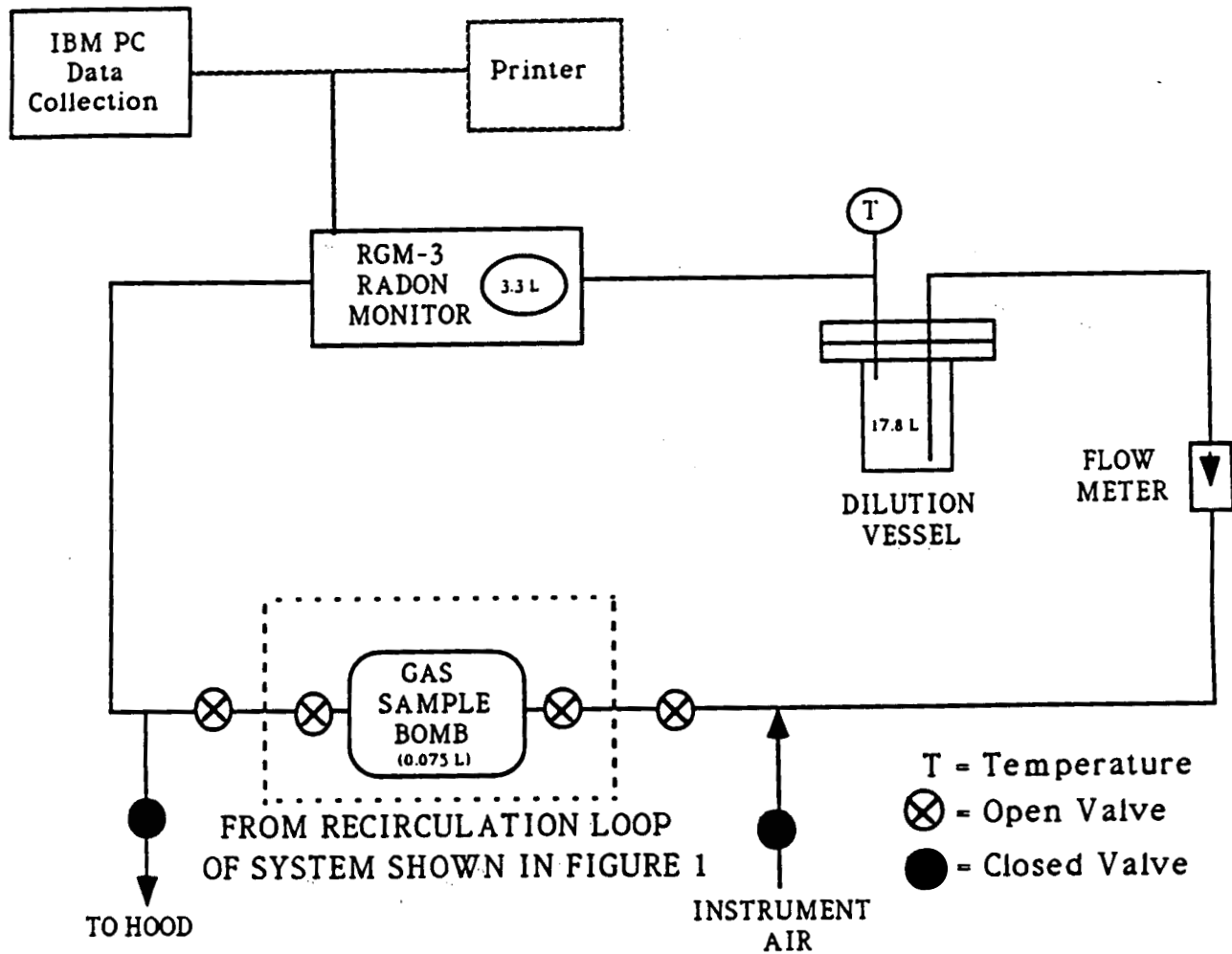
Figure 3. GAS DISCHARGE THROUGH ACTIVATED CARBON
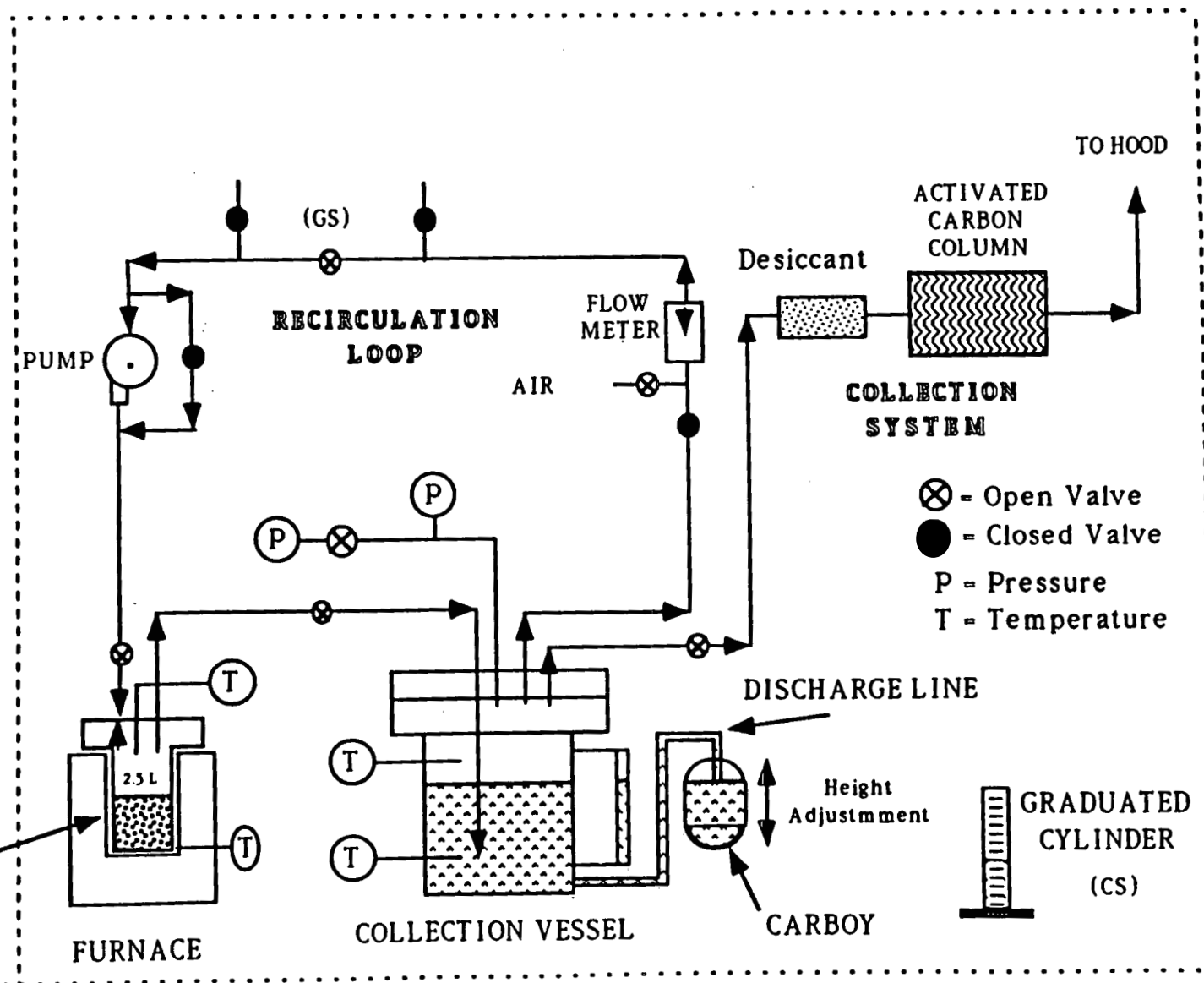
Figure 4. CLOSED SYSTEM FOR ON-SITE RADON MONITORING

RGM-3 RADON MONITOR

T

17.8 L

DILUTION VESSEL

FLOW METER

TO HOOD

INSTRUMENT AIR

2.5 L

CRUCIBLE OF K-65 MATERIAL

⊗ = Open Valve

● = Closed Valve

T = Temperature
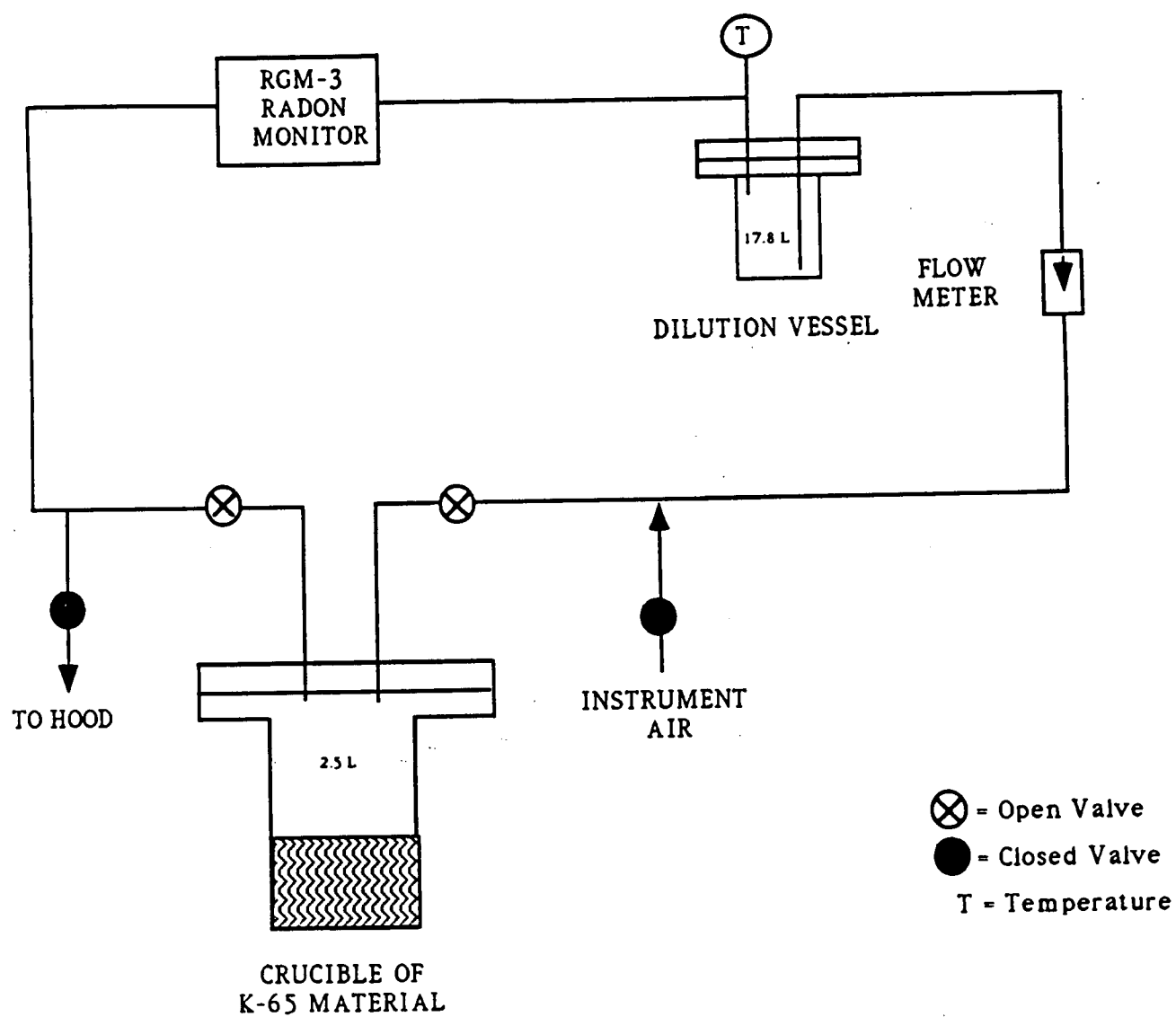
FIGURE 4a. OPEN SYSTEM ON-SITE RADON MONITORING

TO HOOD

OPEN TO
ROOM
AIR

RADON
MONITOR

FLOW
METER

⊗ - Open Valve

CRUCIBLE OF
K-65 RESIDUE
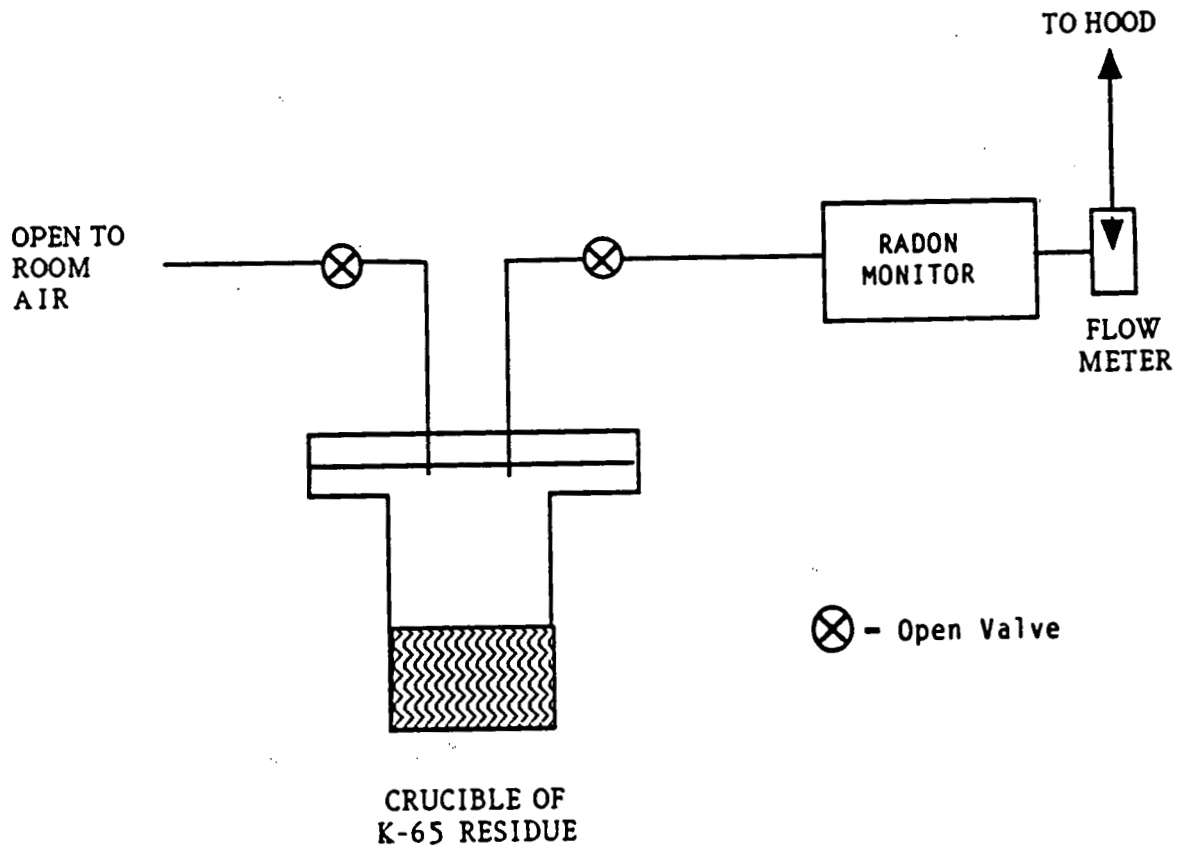
FIGURE 5

Flow Chart of Experimental Method

```
┌─────────────────────────┐    ┌─────────────────────┐    ┌─────────────────────┐    ┌─────────────────────┐
│ 1. Heat Residue and     │    │ 2. Shut Off Power   │    │ 3. Close Valve      │    │ 4. When cool, Set   │
│    Collect Off-Gas      │───▶│    After 2 Hours at │───▶│    Between          │───▶│    Valves           │
│ (See Valve Configuration│    │    1150 C           │    │    Crucible and     │    │    as drawn in      │
│  in Figure 1a)          │    │                     │    │    Collection       │    │    Figure 2         │
└─────────────────────────┘    └─────────────────────┘    │    Vessel           │    └─────────────────────┘
                                                           └─────────────────────┘
```

```
┌─────────────────────────┐    ┌─────────────────────┐    ┌─────────────────────┐    ┌──────────────┐
│ 5. Turn Pump on to      │    │ 6. Turn Pump Off    │    │ 7. Remove Sample    │    │ 8. Repeat    │
│    Obtain               │───▶│    and Close Valves │───▶│    Bomb from        │───▶│    Boxes     │
│    Homogeneous Mixture  │    │    on Sample Bomb   │    │    Recirculation    │    │    5 & 6     │
└─────────────────────────┘    └─────────────────────┘    │    Loop and Place   │    └──────────────┘
                                                           │    in Radon         │
                                                           │    Monitoring Loop  │
                                                           │  (as Shown in       │
                                                           │   Figure 2a).       │
                                                           └─────────────────────┘
```

```
┌─────────────────────────┐    ┌─────────────────────┐    ┌─────────────────────────┐
│ 9. Send Sample Bomb     │    │ 10. When no further │    │ 11. Remove Crucible     │
│    for Chemical Analysis│───▶│     Samples are     │───▶│     from collection     │
│                         │    │     desireed, set   │    │     system (Figure 1)   │
│                         │    │     valves as shown │    │     and place           │
│                         │    │     in Figure 3 and │    │     in Radon Monitoring │
│                         │    │     flush out system│    │     Systems             │
│                         │    │     through         │    │     shown in Figures    │
│                         │    │     activated       │    │     4 & 4a.             │
│                         │    │     charcoal        │    └─────────────────────────┘
└─────────────────────────┘    └─────────────────────┘
```

Phase I - During the first phase of the test, the residue was melted in a 4-inch-diameter by 12-inch-tall (about 2.5 L) inconel crucible in a bench-scale furnace. The off-gas was collected and the volume measured by displacing water from a collection vessel (see Figure 1a). After the crucible cooled, the second phase of the test began.

Phase II - During this phase, a recirculation pump was used to thoroughly mix all of the gas in the system. After the gas was well mixed, samples were collected in a sample bomb (see Figure 2). Some of these were transferred to the monitoring loop and analyzed for radon using an Eberline RGM-3 radon gas monitor (see Figure 2a). Others were sent for chemical analysis.

Phase III - The third phase involved flushing the off-gas through an activated carbon bed that was subsequently analyzed for radon exposure (see Figure 3).

Phase IV - During the final phase, both the vitrified and nonvitrified residue were monitored for radon release using the systems shown in Figures 4 and 4a.

The remainder of this report describes the results of the tests, the methods used to conduct the tests, and the techniques used to measure radon emanation. Appendices A and B, respectively, describe the test procedure for this work and the methods used to test radon behavior.

K-65 VITRIFICATION TESTS: METHODS AND EXPERIMENTAL DATA

CHARACTERIZATION OF K-65 RESIDUE

The K-65 residue was packaged in two Lexan tubes, both labeled Silo 1, NE &
NW, but one labeled BT-3 and the other BT-4. As received, the residue had a high
moisture content (about 29 wt%) and contained rock and other miscellaneous items
(e.g., metal springs, cloth, and leather gloves). To provide a consistent basis, the
residue was dried and sieved through No. 16 (ASTM) mesh to remove the rock and
other items. This mesh has a sieve opening of 1.19 mm and is equivalent to 14 mesh
in the Tyler series. Some of the dried and sieved residue was tamped into a graduated
cylinder and a specific gravity of 1.06 was measured. Prior to vitrification, the residue
was separated into batches and a slurry was prepared using NaOH solution as the glass
former addition. The residue was divided as shown in Table 1.

TABLE 1

Accounting Data for K-65 Residue

Vitrified Material:
Small Test Batch #1	83 g
Small Test Batch #2	164 g
Test #1	968 g
Test #2	500 g
Test #3	946 g
Non-Vitrified Material:	926 g
Rock	1390 g
Glove, Cloth, Springs	83 g
Moisture (29%)	2067 g

Total	7127 g

The K-65 residue has been characterized by various parties. Two different
analyses were performed by PNL, one during 1989 and the other during 1990. Controls
for Environmental Pollution (CEP), Inc. analyzed the residue during 1990, and National
Lead of Ohio (NLO) had previously analyzed the residue. Tables 2 and 3 summarize
the results of the analyses.

TABLE 2

Summary of Chemical Analyses of K-65 Residue:
Not Normalized (expressed in wt% as oxides)

Element	CEP-90 average	PNL-90 average	NLO average	PNL-89 average	Average of averages
Si	52.75%	55.40%	43.59%	53.75%	51.37%
Pb	9.57%	9.01%	5.49%	8.85%	8.23%
Fe	9.01%	4.86%	1.72%	3.75%	4.83%
Ba	7.57%	2.63%	0.08%	5.39%	3.92%
Al	6.46%	3.45%	1.65%	3.11%	3.67%
Ca	1.23%	2.57%	5.44%	2.82%	3.01%
Mg	2.24%	3.47%	1.40%	1.28%	2.10%
Na	1.48%	1.08%	0.94%	1.60%	1.28%
P	0.00%	4.60%	0.00%	0.64%	1.31%
Ti	0.43%	1.34%	0.11%	0.31%	0.55%
K	0.47%	0.51%	0.19%	0.86%	0.51%
Ni	0.64%	0.62%	0.00%	0.44%	0.42%
Co	0.25%	0.37%	0.22%	0.27%	0.28%
Mo	0.24%	0.32%	0.03%	0.20%	0.20%
Cr	0.49%	0.00%	0.02%	0.06%	0.14%
Cu	0.06%	0.11%	0.06%	0.10%	0.08%
Ce	0.00%	0.00%	0.00%	0.30%	0.08%
V	0.12%	0.00%	0.04%	0.13%	0.07%
La	0.00%	0.00%	0.00%	0.26%	0.07%
U	0.00%	0.00%	0.00%	0.16%	0.04%
Mn	0.06%	0.04%	0.03%	0.03%	0.04%
Zr	0.00%	0.00%	0.00%	0.15%	0.04%
Nd	0.00%	0.00%	0.00%	0.12%	0.03%
Sr	0.00%	0.00%	0.00%	0.11%	0.03%
Be	0.02%	0.00%	0.00%	0.09%	0.03%
Th	0.00%	0.00%	0.00%	0.06%	0.02%
Sn	0.04%	0.00%	0.01%	0.00%	0.01%
Se	0.00%	0.04%	0.00%	0.00%	0.01%
Zn	0.02%	0.01%	0.00%	0.00%	0.01%
Au	0.00%	0.009%	0.00%	0.00%	0.002%
Total	93.17%	90.42%	61.03%	84.70%	82.33%

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TABLE 3

Summary of Chemical Analyses of K-65 Residue:
Normalized (expressed in wt% as oxides)

Element	CEP-90 average	PNL-90 average	NLO average	PNL-89 average	Average of averages
Si	56.62%	61.28%	71.43%	63.46%	63.20%
Pb	10.27%	9.97%	9.00%	10.45%	9.92%
Fe	9.67%	5.37%	2.81%	4.43%	5.57%
Ba	8.12%	2.90%	0.13%	6.36%	4.38%
Al	6.94%	3.81%	2.71%	3.67%	4.28%
Ca	1.32%	2.85%	8.91%	3.33%	4.10%
Mg	2.41%	3.83%	2.30%	1.51%	2.51%
Na	1.59%	1.19%	1.55%	1.89%	1.55%
P	0.00%	5.08%	0.00%	0.76%	1.46%
Ti	0.46%	1.48%	0.18%	0.37%	0.62%
K	0.50%	0.56%	0.32%	1.02%	0.60%
Ni	0.68%	0.69%	0.00%	0.52%	0.47%
Co	0.27%	0.41%	0.36%	0.32%	0.34%
Mo	0.26%	0.35%	0.05%	0.24%	0.22%
Cr	0.53%	0.00%	0.03%	0.07%	0.16%
Cu	0.06%	0.12%	0.10%	0.12%	0.10%
Ce	0.00%	0.00%	0.00%	0.35%	0.09%
V	0.13%	0.00%	0.06%	0.15%	0.09%
La	0.00%	0.00%	0.00%	0.31%	0.08%
U	0.00%	0.00%	0.00%	0.19%	0.05%
Mn	0.06%	0.04%	0.04%	0.04%	0.04%
Zr	0.00%	0.00%	0.00%	0.18%	0.04%
Nd	0.00%	0.00%	0.00%	0.14%	0.04%
Sr	0.00%	0.00%	0.00%	0.13%	0.03%
Be	0.02%	0.00%	0.00%	0.11%	0.03%
Th	0.00%	0.00%	0.00%	0.07%	0.02%
Sn	0.04%	0.00%	0.02%	0.00%	0.02%
Se	0.00%	0.04%	0.00%	0.00%	0.01%
Zn	0.02%	0.01%	0.00%	0.00%	0.01%
Au	0.00%	0.01%	0.00%	0.00%	0.003%
Total	100.00%	100.00%	100.00%	100.00%	100.00%

Extraction procedure (EP) toxicity data were obtained for the K-65 residue (Buelt, 1989). The data are given in Table 4. All of the heavy metals from the EP toxicity list (Ag, As, Ba, Cd, Cr, Hg, and Se), with the exception of lead, were below the regulatory limit. Table 5 summarizes results from a gamma scan conducted to determine the isotopic content of the residue. Isotopes other than those listed in Table 5 may have been present.

TABLE 4

EP Toxicity Data for Nonvitrified K-65 Residue (Buelt, 1989)

Metals	Value(mg/L)	Limit(mg/L)
Arsenic	<1.0	5.0
Barium	0.76	100
Cadmium	0.1	1.0
Chromium	<0.2	5.0
Lead	630[a]	5.0
Mercury	<0.03	0.2
Selenium	<0.1	1.0
Silver	<0.1	5.0

(a) Value for lead was above the regulatory limit

TABLE 5

Isotopic Content of K-65 Residue

| Isotope	(nCi/g)
Th-230	264.7
Ra-226	479.4
Pb-214	297.8
Bi-214	280.0
Pb-210	338.2
Pa-231	29.8
Th-227	21.1
Ra-223	22.4
Rn-219	20.8
Pb-211	65.7

DESCRIPTION OF EXPERIMENTAL SYSTEM

Phase I - The experimental system, as shown in Figure 1, was designed to contain and cool all of the off-gas produced during vitrification of the slurry contained in the crucible. As the crucible was heated (see the heating schedule shown in Figure 6), the gas already present in the system expanded and an off-gas was produced from the decomposition of the K-65 residue. These gases then bubbled through and displaced the water in the collection vessel, and the displaced water was collected in a carboy (see Figure 1a). The use of both water displacement and adjustments to the height of the carboy ensured that the pressure in the system was essentially equal to the ambient pressure. At the conclusion of the heating period the valve between the crucible and the collection vessel was closed, and the valves to the recirculation loop were opened so that as the gas cooled and contracted, the water in the collection vessel was not sucked back to the crucible through the dip leg. Instead, the gas traveled through the recirculation loop, and the water in the carboy flowed back into the collection vessel as needed. After the system cooled down, the volume of the off-gas produced was obtained by measuring the volume of displaced water.

Phase II - The next phase involved collecting samples of the off-gas and measuring the radon concentration. This was done by first recirculating the off-gas through the recirculation loop to ensure that a homogeneous mixture was sampled. Samples were then collected in 75-mL sample bombs as shown in Figure 2. An Eberline RGM-3 radon gas monitor was purchased for monitoring the gas samples on-site, as shown in Figure 2a. The sample in the gas bomb was diluted with 21.1 L of air to keep the concentration of radon within the operating range of the radon monitor.

Phase III - The remaining off-gas was then passed through an activated charcoal column (see Figure 3), which collected essentially all of the radon. This procedure provided back-up data to that obtained with the radon gas monitor.

Figure 6
Typical Heating Schedule
(Data from Test #2)

TEMPERATURE (C)

ELAPSED TIME (MIN)

16

25

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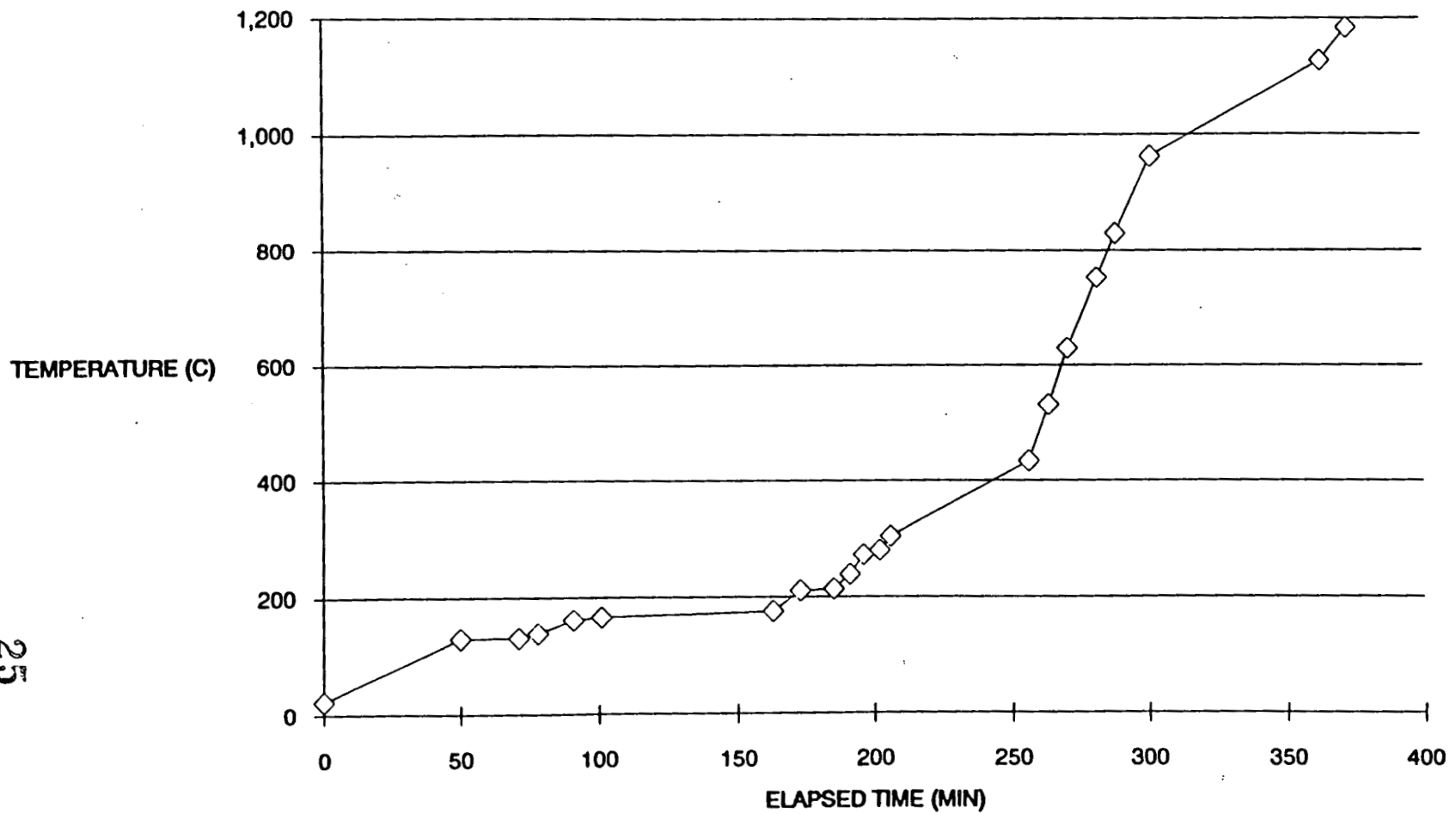
Phase IV - For the final phase, the radon emanating from the vitrified K-65 residue was measured using the system shown in Figure 4 and Figure 4a. Prior to beginning these measurements, the systems were flushed with air to provide a 0 pCi/L starting point. The complete test plan is included as Appendix A.

TEST #1

The slurry used in the first test consisted of 968 grams of K-65 residue, 166 grams of reagent grade NaOH, and 885 grams of water. Based on calculations, this combination would produce a glass that is 12 wt% Na_2O and a slurry that was 44 wt% water. The slurry was quite dilute and probably contained more water than would be used during actual processing conditions.

While this batch was processed, the boiling rate was excessive and carried over about 170 grams of the solid K-65 residue into the collection vessel. Also during this test, more off-gas was produced (13.5 liters) than the system could contain. Therefore, 8.4 liters of off-gas was vented to the activated charcoal collection column to avoid displacing all of the water from the collection vessel and, consequently, losing track of the quantity of off-gas generated.

After the system cooled down, off-gas samples were collected for chemical analyses and for measurements of radon concentration. The results of the chemical analyses are given in Table 6. In addition, samples of the vitrified residue were analyzed for percent composition and tested using the Toxicity Characteristic Leachate Procedure (TCLP). The results are presented in Tables 7 and 8. Based on the data in Table 8, the vitrified K-65 residue is nonhazardous as tested by the TCLP, whereas the nonvitrified K-65 residue is hazardous because it exceeds the EP Toxicity limit for lead (see Table 4). A specific gravity of 3.1 was measured for the vitrified residue. Table 9 compares the specific gravity of the nonvitrified residue with the vitrified residue.

TABLE 6

Volume Percent Composition of Vitrification Off-Gas

Component	Vol%
Nitrogen	74.3
Oxygen	19.0
Carbon Dioxide	5.50
Argon	0.88
Hydrogen	0.26
Methane	0.04
Helium	0.01
Other	0.01

TABLE 7

Weight Percent Composition (as oxides) of Vitrified K-65 Residue

Oxide	Wt%
Si	52.75
Na	13.49
Pb	9.57
Fe	9.01
Ba	7.57
Al	6.46
Mg	2.24
Ca	1.23
Ni	0.64
Cr	0.49
K	0.47
Ti	0.43
Co	0.25
Mo	0.24
V	0.12
Cu	0.06
Mn	0.06
Sn	0.04
Zn	0.02
Be	0.02
Sb	0.02
Cd	0.01
Ag	0.00
Hg	0.00

TABLE 8

TCLP Leachate Results for Vitrified K-65 Residue
(Concentration of Metals in Leachate)

Metal	Value (mg/L)	Limit (mg/L)
Arsenic	<0.01	5.0
Barium	0.98	100.0
Cadmium	<0.05	1.0
Chromium	<0.05	5.0
Lead	0.30	5.0
Mercury	<0.0004	0.2
Selenium	<0.01	1.0
Silver	<0.05	5.0

TABLE 9

Specific Gravity of Residue

Material	SpG
Nonvitrified Residue	1.06
Vitrified Residue	3.1

TEST #2

For the second test, a smaller batch of slurry was prepared so all of the off-gas could be collected. This batch consisted of 500 grams of K-65 residue, 72 grams of NaOH, and 114 grams of water. Based on calculations, this combination would produce a glass that was 10 wt% Na_2O. The quantity of water added was just enough to make a thick slurry, and all of the off-gas (7.3 liters) produced during this run was collected without interim venting. The composition of the off-gas is given in Table 10. A condensate sample from the collection vessel was also analyzed; those results are given in Table 11.

TABLE 10

Volume Percent Composition of the Vitrification Off-Gas

Component	Vol%
Nitrogen	77.2
Oxygen	17.1
Other Ions	3.34
Water	1.4
Argon	0.9
Carbon Dioxide	0.06
Organic Compounds	None detected

TABLE 11

Composition of Condensate

Component	(mg/L)
Aluminum	<0.1
Antimony	0.04
Arsenic	<0.01
Barium	0.07
Cadmium	<0.01
Calcium	14.1
Chromium	<0.01
Cobalt	0.01
Iron	0.06
Lead	0.09
Magnesium	3.7
Mercury	0.0029
Nickel	0.1
Phosphorus	0.2
Potassium	0.7
Selenium	0.19
Sodium	3.2
Uranium	0.011

20

TEST #3

For the third test, the remaining K-65 residue was divided. Half of the residue was used for the third melt, and the other half was used to obtain emanation measurements from the nonvitrified residue. These batches had a mass of 946 grams and 926 grams, respectively. The residue was slurried using 149 grams of NaOH and 341 grams of water. This combination was calculated to produce a glass that was 11 wt% Na_2O. Because of the large batch size, 4.5 liters of off-gas were vented during the vitrification process to avoid total displacement of the water in the collection vessel. The total volume of off-gas produced was 12.2 liters.

RADON MONITORING

RADON RELEASED DURING VITRIFICATION

As described previously, samples of the vitrification off-gas were collected and diluted to determine the radon content. In addition, the off-gas was flushed through the activated charcoal column which was analyzed for radon exposure. The results of all these measurements are given in Table 12. Some of the data may have been affected by the use of a pump which required oil for operation. Later tests revealed a strong tendency for oil to absorb radon.

Table 12

Radon Released During Vitrification Process

Test #	K-65 (grams)	Radon on Charcoal (μCi)	Radon measured with RGM-3 (μCi)
1	970	72	--
1	970	30	60
2	500	0.24	0.32
3	946	--	8.7

For Test #1, two separate activated charcoal columns were used. The first column was used to collect the radon from the off-gas vented during processing. The 72 μCi were collected from 8.4 liters of off-gas, which means that the concentration was equal to 8.6 μCi/L. (Note: The pump was not used to obtain this data point.) Adding the 30 μCi from the second column results in a total of 102 μCi. This represents only 23% of the 445 μCi present when this quantity of residue reaches secular equilibrium. The second column was used to collect all of the radon in the system as it was flushed out. The radon measurements differed between tests, but there is fair agreement between the data obtained with the radon monitor and the activated charcoal columns.

The ability of materials to retain radon was examined by Barretto (Barretto, 1975). In his studies, various minerals were heated to 1200 °C. The radon emanation decreased by 80% to 90% as pores and cracks in the minerals "healed." Thus, as the K-65 residue is heated, the emanation rate probably decreases by the same mechanism. Diffusion of gases through glass at room temperature is almost nonexistent, as mentioned previously. For 930 grams of K-65 residue, the volume of radon gas at secular equilibrium is so small (2.92E-10L) that bubbles will not form. Even if bubbles could form, their removal from molten glass (one of the common processing challenges faced by the glass industry) is not likely because the viscosity of molten glass is typically about 10,000 cp. It is also probable that this trace quantity of radon is easily dissolved in the glass.

The small percentage of released radon could be managed easily with activated charcoal beds. For a 10 ton/day melter, a 2500-liter charcoal bed could absorb all of the radon produced during a 40-day period (Fusamura, et al., 1963). Two of these beds could be employed so that as one was absorbing radon, the other would be regenerated by taking it off-line and allowing the absorbed radon to decay away.

CLOSED SYSTEM TESTS

The glass produced during Test #2 was monitored for radon emanation using the system shown previously (see Figure 4). Over a 39-hour period (data points were taken every hour), the radon concentration gradually increased from 0 pCi/L (the system was initially flushed with air) to about 103 pCi/L in a volume of 21.4 liters (see Figure 7). The radon emanating from the nonvitrified K-65 residue was also measured. The residue was placed in a crucible in the system shown in Figure 4, filling 4 to 5 inches of the 12-inch-tall crucible. The amount of radon emanating from the nonvitrified residue was so great that it exceeded the range limitations of the RGM-3 radon monitor (100,000 pCi/L) within 6 hours, as shown in Figure 8.

The volume of the system used was 21.4 liters, so a total of 2.1 μCi of radon was present after the 6 hours. Since the residue theoretically generates 3.4 million pCi/hr, the net radon emanation rate is a fraction of the total generated in the residue. Much of the radon is absorbed by the residue, trapped within crystalline structures, or decays before it can diffuse to the air over the residue. The amount of radon that does emanate depends on these factors, and the amount of diffusion and absorption of radon depends on the radon concentration present in the system. Freeman (Freeman, 1981), who also references Nielson (Nielson, et al., 1981), acknowledges that radon emanation is reduced as its concentration builds above the emanating source. The data from the closed system does not provide enough information for the emanation rate to be determined. However, if the radon concentration had not exceeded the limitations of the radon monitor, the residue could have been monitored continuously until secular equilibrium was reached. At secular equilibrium the emanation rate would equal the decay rate measured by the radon monitor.

Measuring these very high concentrations was difficult since the instruments available were designed for handling commonly observed environmental concentrations which are much lower. To get a measurement of the radon concentration over the nonvitrified residue, an attempt was made to collect a sample in a gas bomb, and then dilute it in the radon monitoring loop (see Figure 2a). However, to collect a sample, the recirculation pump was again used. It was during these attempts that it became obvious that the pump may have been absorbing radon.

Figure 7. Measured Radon Concentration in Closed System for Vitrified K-65 Residue from Test # 2
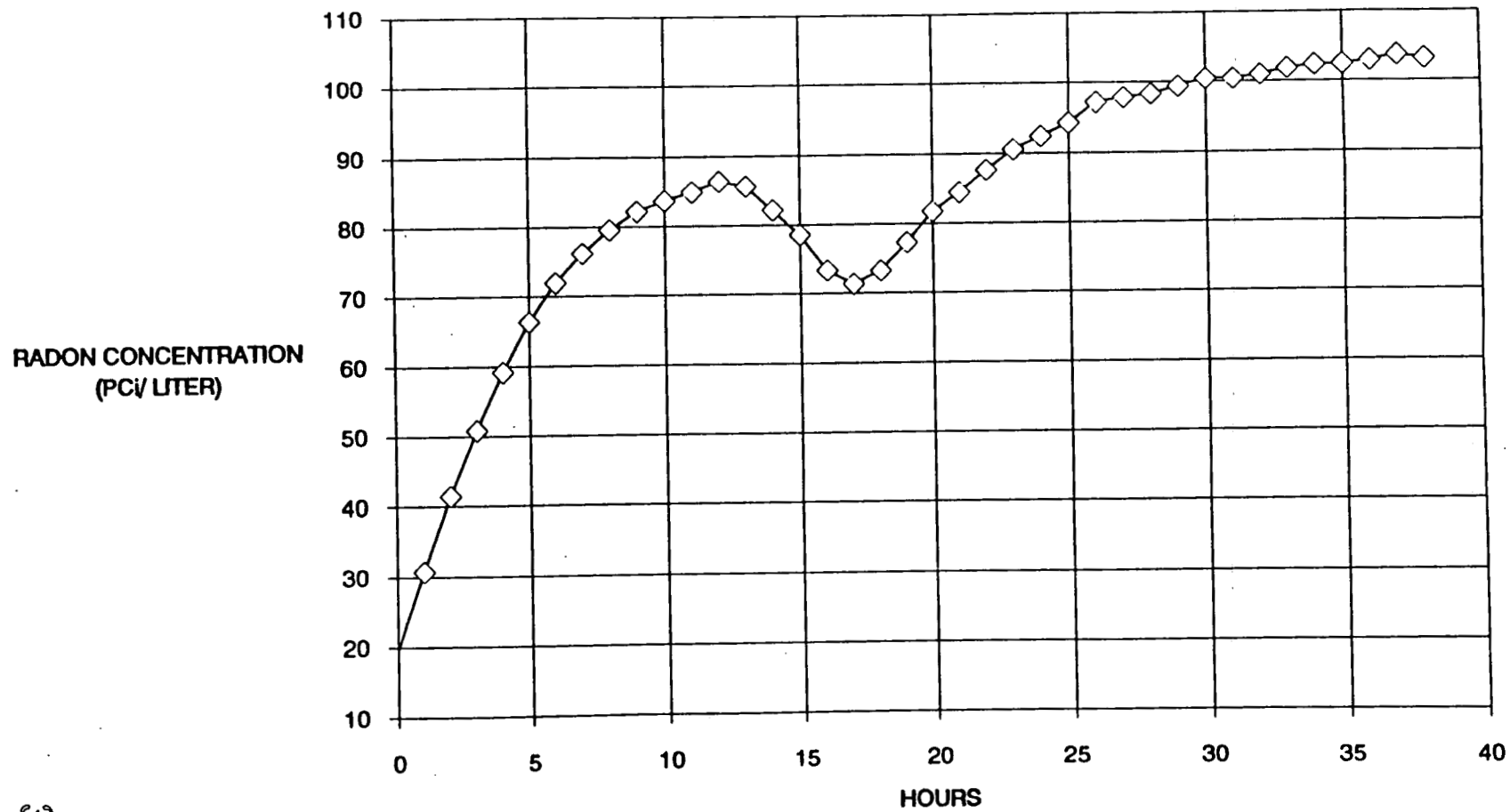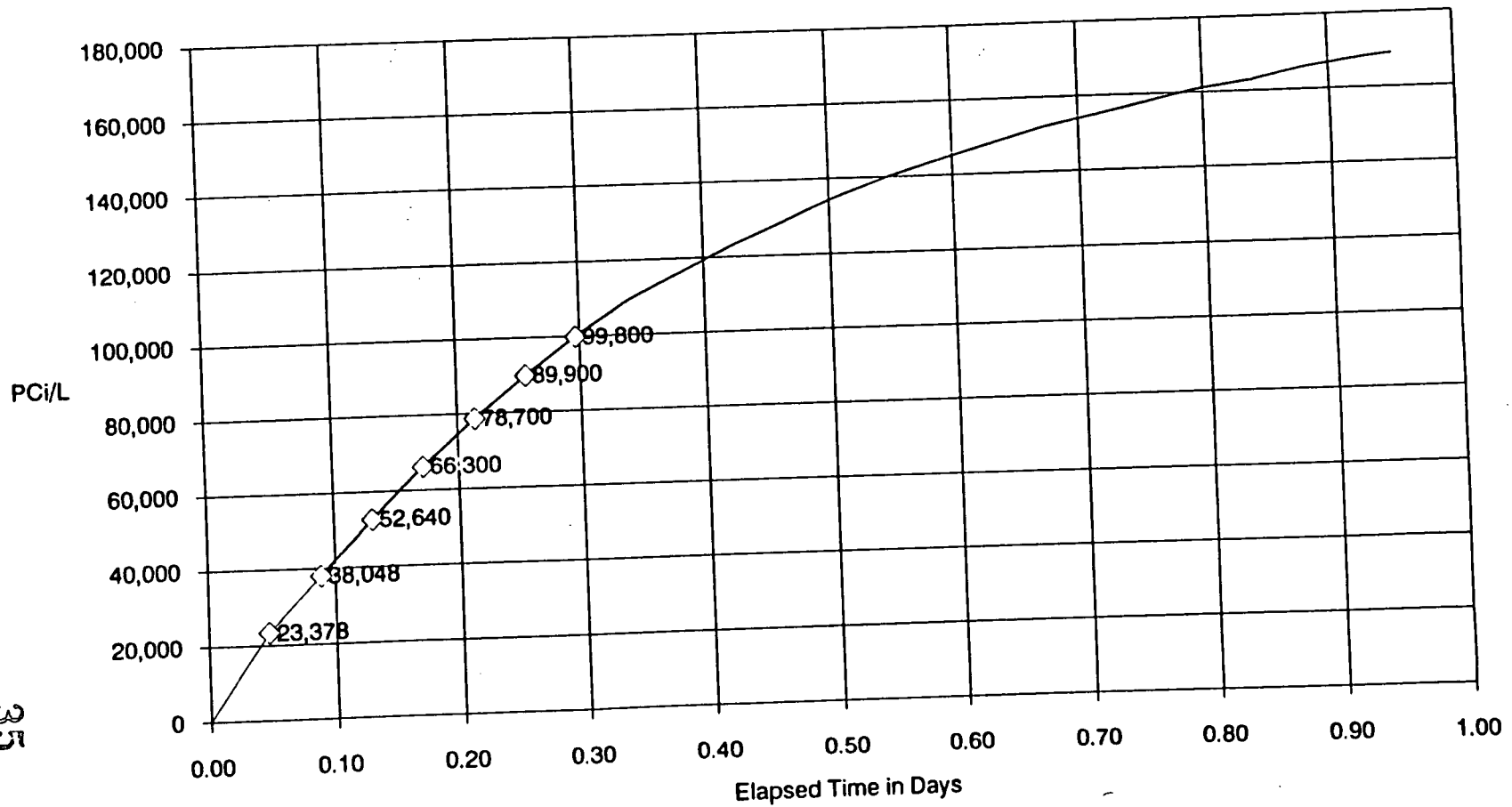
RADON CONCENTRATION
(PCi/ LITER)

HOURS

Figure 8. Measured Radon Concentrations in Closed System for Non-Vitrified K-65 Residue
(Data Points Taken Every Hour)

PCi/L

Elapsed Time in Days

OPEN SYSTEM TESTS

To avoid high radon concentrations and any concentration-dependent interferences, the radon monitoring configuration was changed from a continuously recirculating closed system to a once-through open system (see Figure 4a). The radon concentration in this test arrangement began at 0 pCi/L, and then reached and maintained a nearly steady value of 3,200 pCi/L for over 420 hours (see Figure 9). At 8 L/min, this represents an emanation rate of 1.5 million pCi/hr or 52,400 pCi/m²/s. This emanation rate accounts for about 50% of the total amount of radon theoretically generated by the decay of the radium-226 present, and is higher than any other rate measured. Figure 10 compares the total amount of radon that would be present at a constant emanation rate of 1.5 million pCi/hr to that measured in the closed system.

We obtained a Pylon AB-5 monitor for parallel monitoring of both the vitrified and non-vitrified residue and for use as a crosscheck between monitors. The Pylon monitor differs from the RGM-3 in the rate at which it pumps. The typical flow rate for the RGM-3 is 8 liters per minute, while the Pylon pumps at less than 0.5 liters per minute. The RGM-3 had experienced high background readings from the high concentrations of radon. Since the Pylon monitor had not accumulated any background, it was used to monitor the vitrified residue from Test #3, and the RGM-3 was used to monitor the nonvitrified residue.

The 946 grams of vitrified K-65 residue from Test #3 occupied about 1.5 inches of the 12-inch-tall crucible. The results from monitoring this residue in a closed-loop system (see Figure 4) are shown in Figure 11. The radon level peaked early in the monitoring period, and then slowly decayed to a nearly constant value.

To provide a common basis for comparing the emanation rate of vitrified and nonvitrified K-65 residue, the vitrified K-65 residue was also measured in an open system, as illustrated in Figure 4a. The concentration of radon in the sweep air going through the canister of vitrified residue is presented in Figure 12. These results indicate

Figure 9. RADON CONCENTRATION IN SWEEP AIR
Non-Vitrified K-65 Residue, Open System

Radon Concentration (PCi / Liter) vs Elapsed Monitoring Time (hrs)
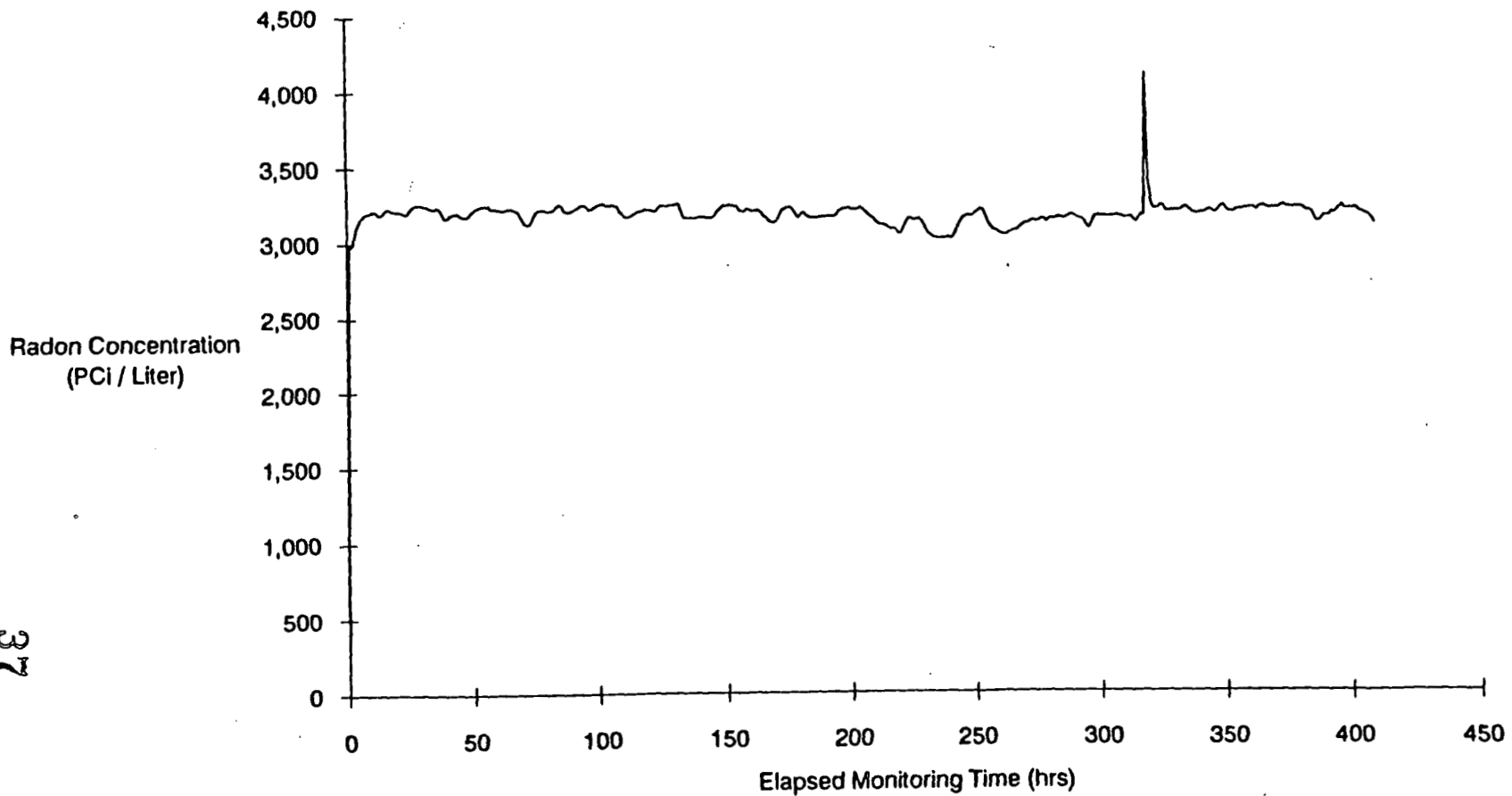
Figure 10. Comparison of Measured and Projected Radon Released from Non-Vitrified K-65 Residue, Extended Projection Accounts for Radon Loss by Decay

Projected at the 1,500,000 pCi/hr measured in the open system

Measured from the closed system

Measured (same data as Figure 7)

PCi

Elapsed Time in Days

Figure 11. MEASURED RADON CONCENTRATION IN GAS PHASE
Vitrified K-65 Residue -- Closed System

RADON
CONCENTRATION
(PCi / Liter)

HOURS

30

39

2830

Figure 12. Radon Concentration in Sweep Air
for Vitrified K-65 Residue in Open System (pCi/Liter)

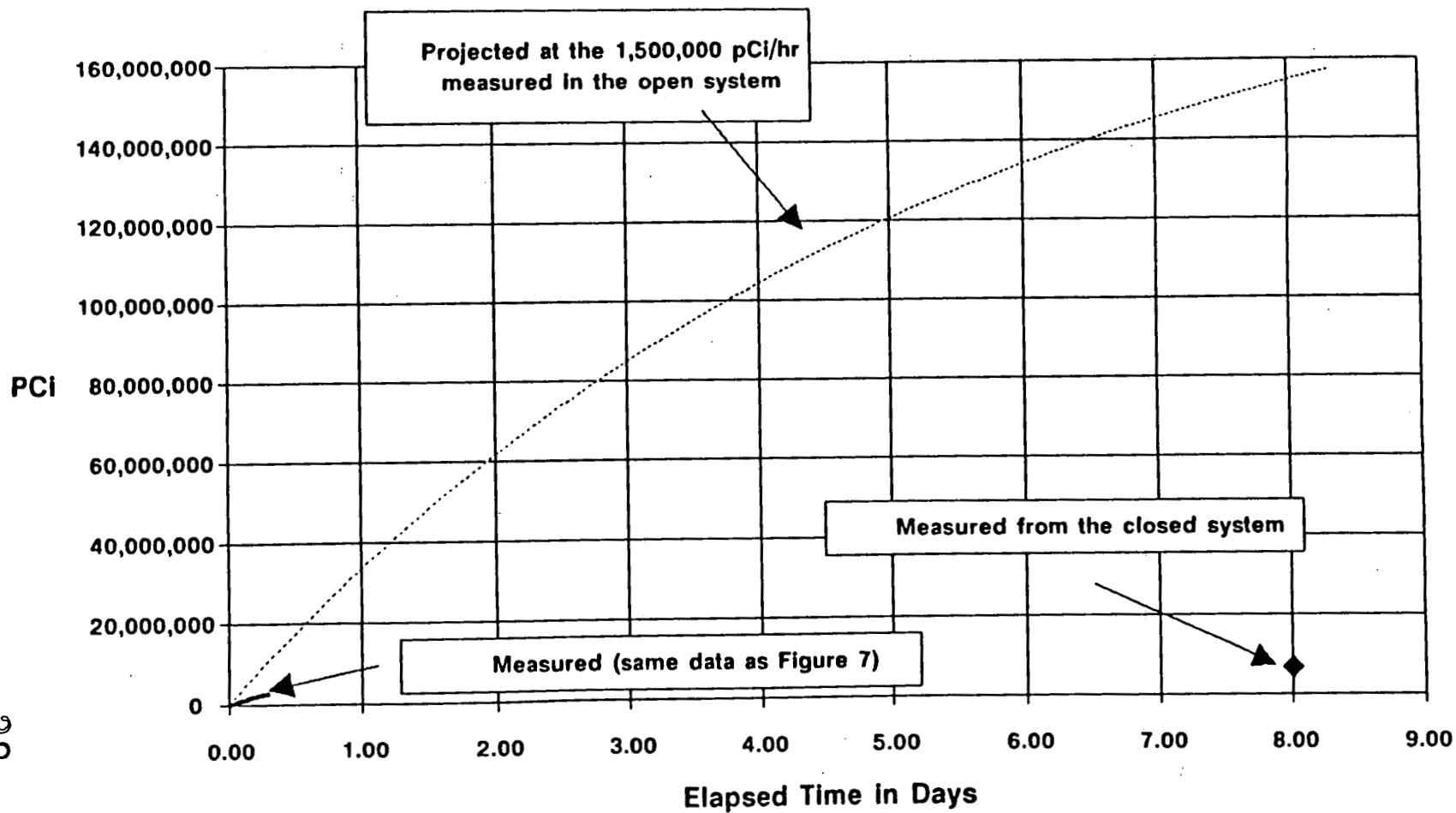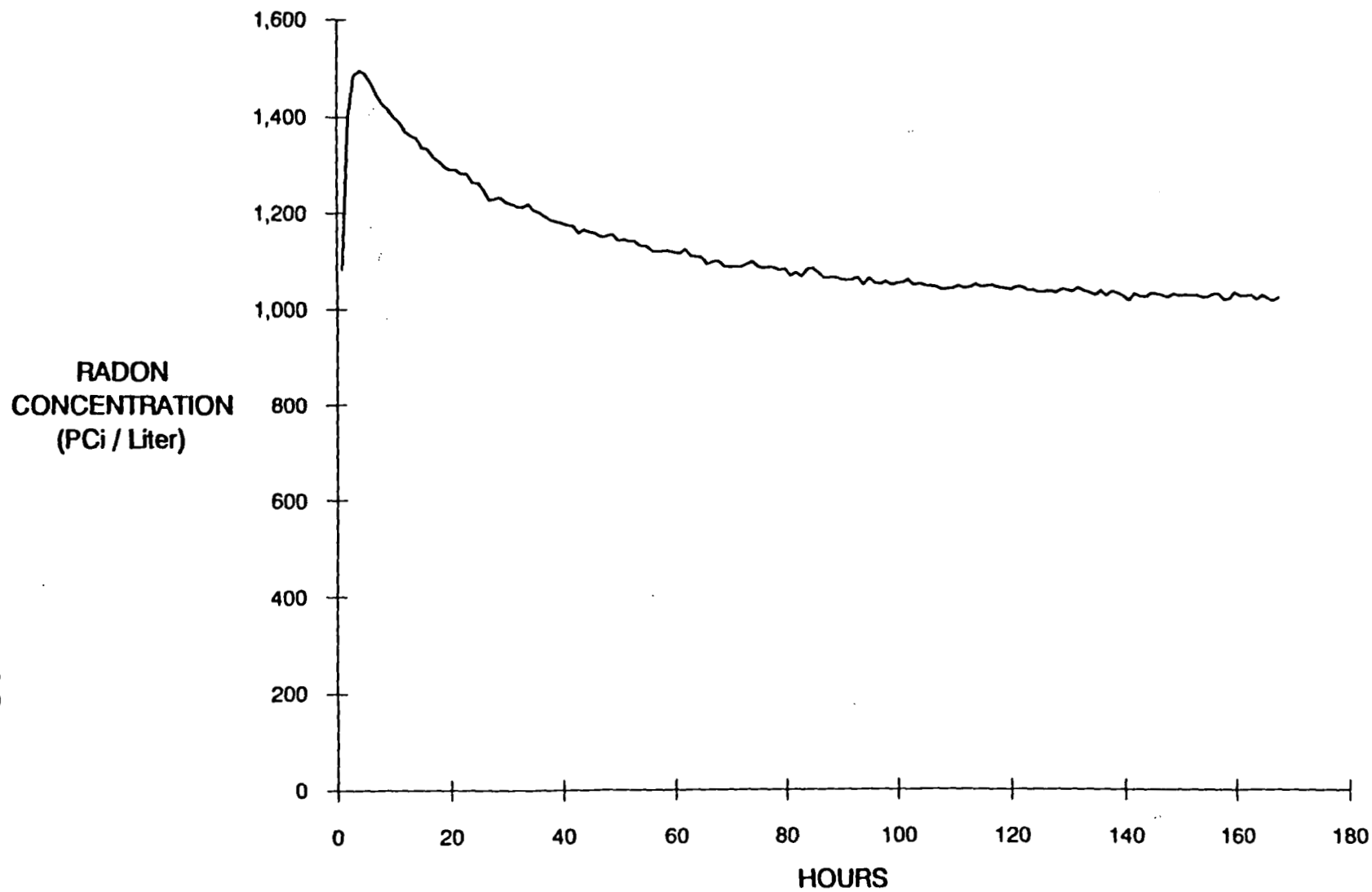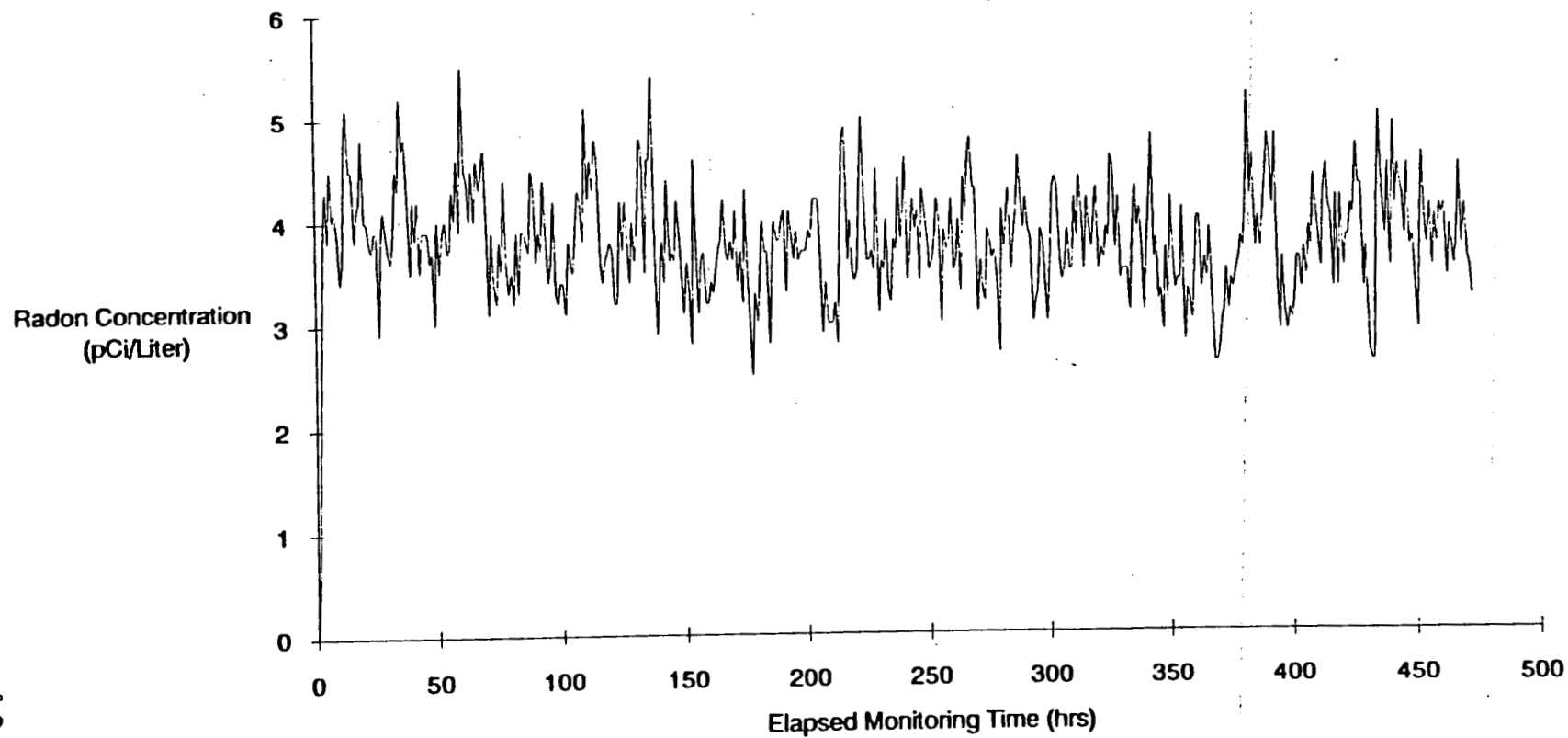that the radon concentration, which began at 0 pCi/L, averaged about 4 pCi/L. For the arrangement used, this represents an emanation rate of 48 pCi/hr, or 1.56 pCi/m²·s. This is over 33,000 times less than the rate for the nonvitrified residue, and is an order of magnitude lower than the EPA limit of 20 pCi/m²·s.

THORON EMANATION FROM NONVITRIFIED K-65 RESIDUE

Measurements to detect the presence of the short-lived radon isotope radon-220, also known as thoron, were made. This isotope of radon has a half life of less than a minute. To determine whether or not thoron was present, the Pylon monitor was used to withdraw a slip stream from the sweep stream pumped by the RGM-3 (see Figure 13; also see Appendix B). After achieving a steady flow through the system, the pump was stopped and the gas in the chamber of the monitor counted. A total count for each minute was taken for 10 consecutive minutes. Three replicates of this technique were performed; the resulting data are presented in Figure 14. The drop in the total counts between the first and second minutes indicates that some thoron is present. However, after the first two readings, the radon daughters began to build and the drop due to thoron was no longer obvious. Although some thoron was present, it certainly was not dominant.

ABSORPTION OF RADON BY VACUUM PUMP OIL

Tests were also conducted to assess absorption of radon by the vacuum pump oil. For these tests, a filter saturated with the oil was placed between the K-65 residue (radon source) and the radon monitor, as shown in Figure 15 (also see Appendix B).

Two separate tests were conducted using two types of oil. For the first test, Dow Corning 704 Diffusion Pump Fluid (pure tetramethyltetraphenyltrisiloxane) was used. Because of the pressure drop across the oil-laden filter, the flow rate through the system was reduced to 6.5 L/min. At this flow rate, the highest concentration of radon measured was 2035 pCi/L. This is equivalent to 13,230 pCi/min. The filter was

41

FIGURE 13

THORON MONITORING SYSTEM

TO HOOD

TO HOOD

OPEN TO
ROOM AIR

RGM-3
RADON
MONITOR

FLOW
METER

CRUCIBLE OF NON-VITRIFIED
K-65 RESIDUE

PYLON
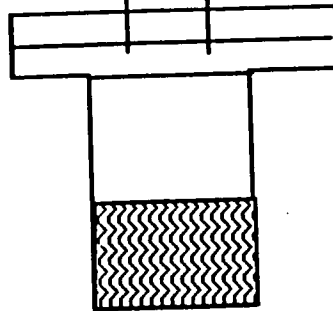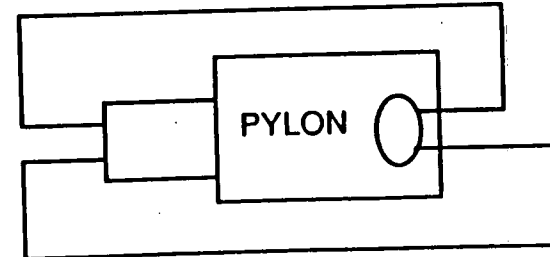
FLOW
METER

Figure 14. Measurements for Thoron Concentration
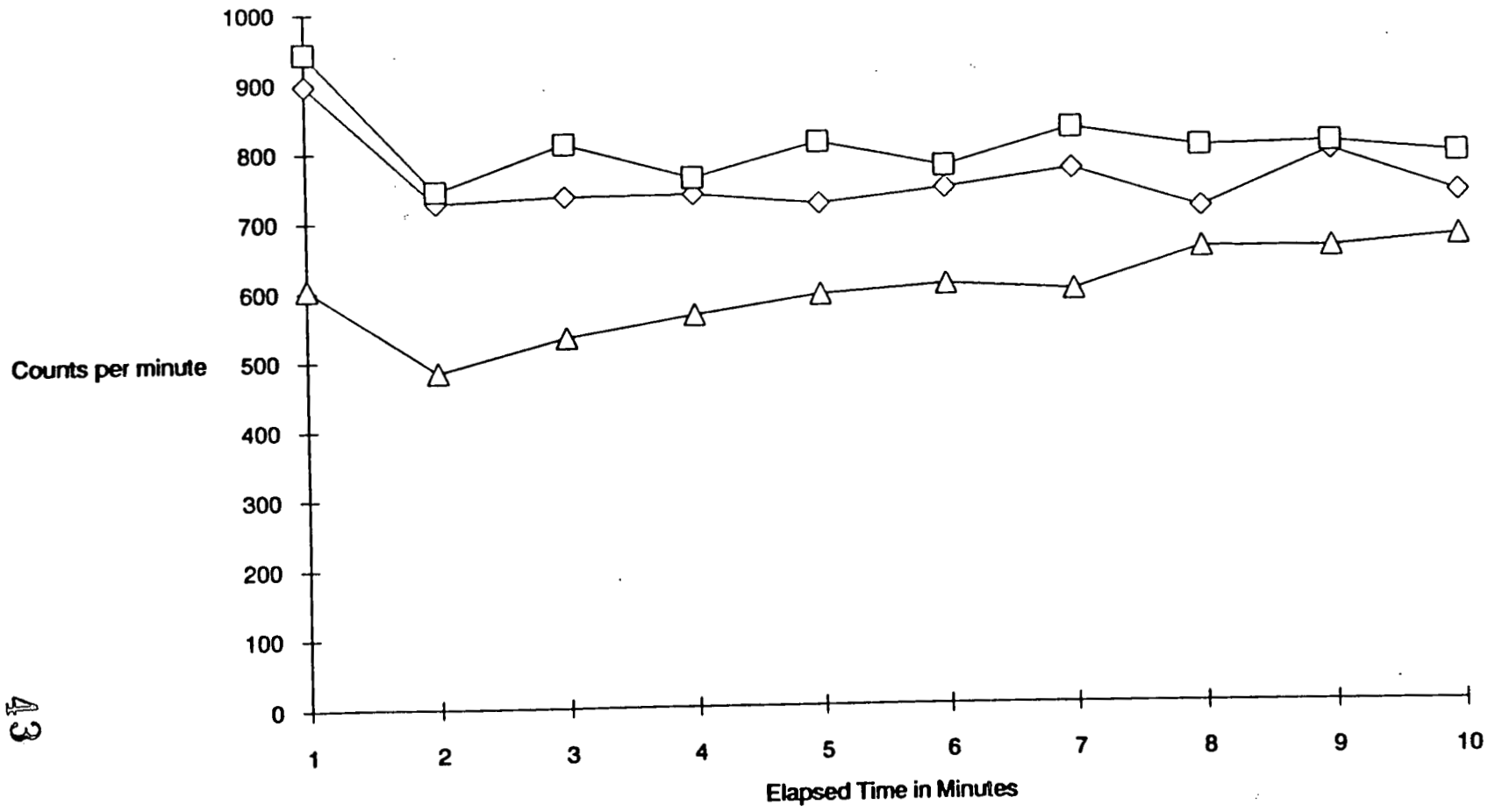
FIGURE 15

OIL ADSORPTION RADON MONITORING SYSTEM

CRUCIBLE OF NON-VITRIFIED
K-65 RESIDUE
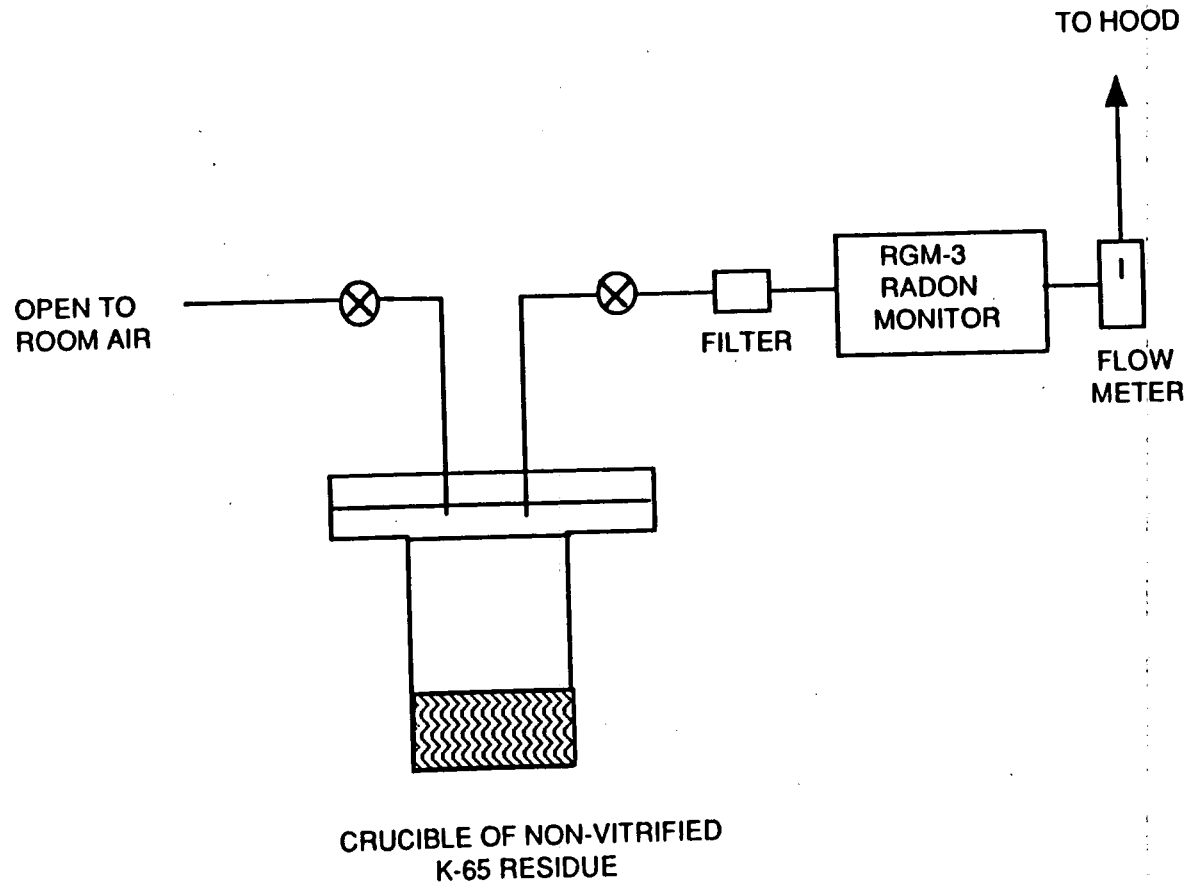
quickly removed from the system after taking this measurement, and then a reading of 3410 pCi/L was obtained at a flow rate of 7.9 L/min. This is equivalent to 26,940 pCi/min. Assuming that the residue is a steady source of radon, the data show that, with a very short contact time of about 0.006 seconds in the filter media, over 50% of the radon was absorbed.

Consolidated Vacuum Corporation's Convoil-20 Vacuum Pump Fluid was used as the oil for the second test. This is a distilled hydrocarbon fluid that is 70% paraffinic mineral oil and 30% napthalic mineral oil. At a flow rate of 6.5 L/min, the radon concentration was 2420 pCi/L. This is equivalent to 15,740 pCi/min and indicates that over 40% of the radon is absorbed during the same short contact time as above.

Based on these results, it seems very likely that purposefully increasing the contact time by using a packed column or some other technique would result in almost complete absorption of the radon.

RADON EMANATION DEPENDENCE ON CONCENTRATION

The radon concentration over the K-65 residue was allowed to build in a closed system for 8 days, at which time a 75-mL sample was diluted with 22.4 liters of air. (see Figure 16; also see Appendix B). The radon concentration measured was 13,150 pCi/L. Therefore, the radon concentration over the K-65 residue was 3.9 million pCi/L (total of 6.7 μCi) after the 8 days. As shown in Figure 10, this value is far below that obtained by a constant emanation rate of 1.5 million pCi/hr. The data indicate that the net emanation rate at higher concentrations is much lower than it is at 3200 pCi/L.

FIGURE 16
Radon Monitoring System

Nonvitrified K-65 Residue
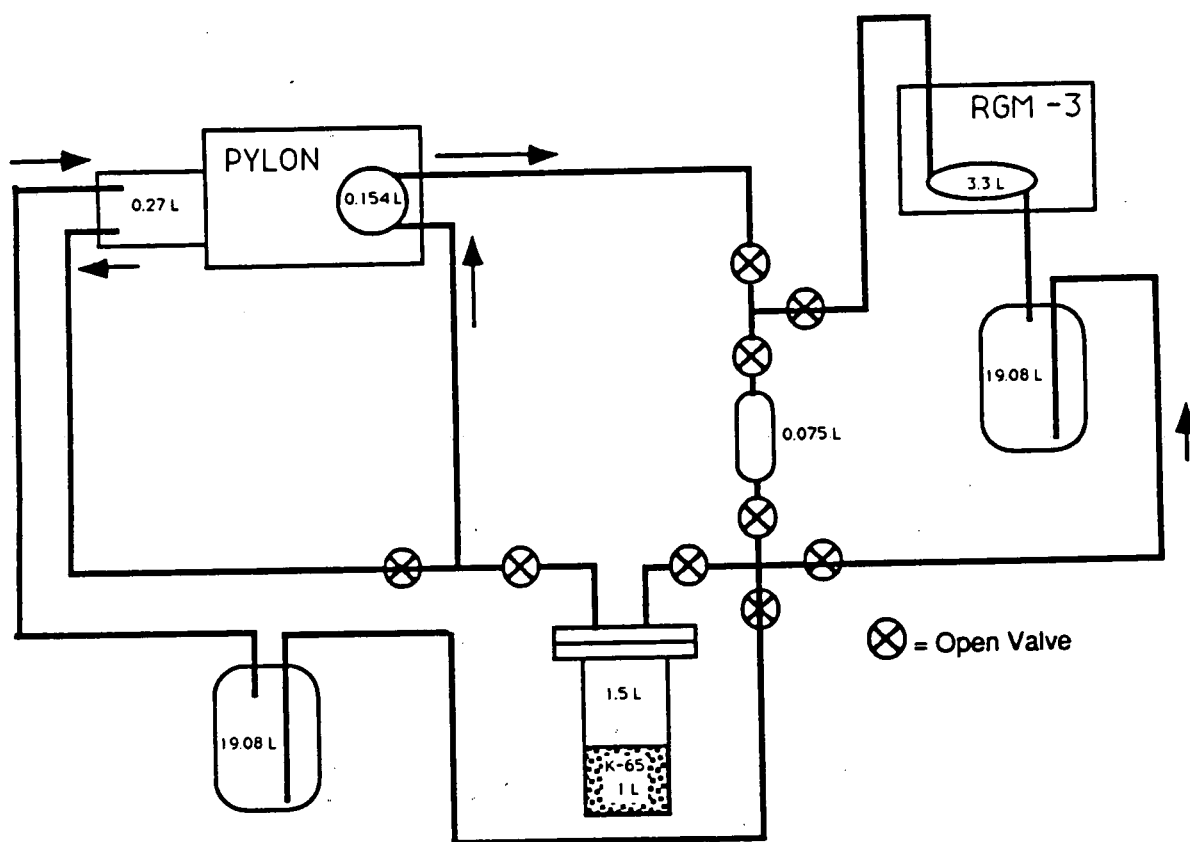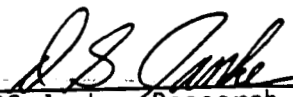
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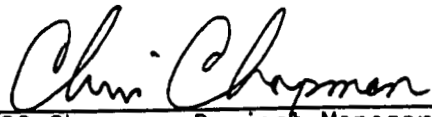
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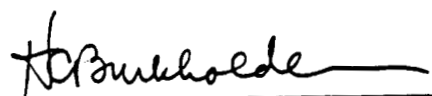
APPENDIX A
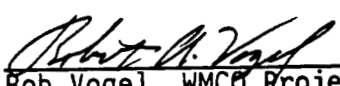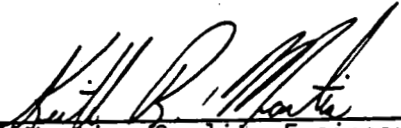
TEST PLAN FOR K-65 OFF-GAS CHARACTERIZATION
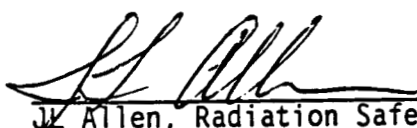
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             DS Janke, Research Engineer   Date

Approved by: _____   8-3-90
             CC Chapman, Project Manager   Date
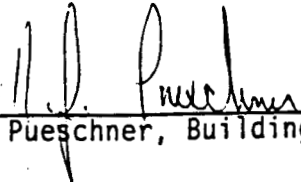
             _____   8/3/90
             HC Burkholder, Section Manager   Date

             _____   8/8/90
             Bob Vogel, WMCO Project Manager   Date

             _____   8/3/90
             KR Martin, Quality Engineer   Date

             _____   8/7/90
             JL Allen, Radiation Safety   Date

             _____   8/8/90
             DW Wright, Industrial Health & Safety   Date

Concurred by: _____   8-3-90
             MJ Pueschner, Building Manager   Date

TEST PLAN FOR K-65 OFF-GAS CHARACTERIZATION

INTRODUCTION

The K-65 Silos at the Feed Materials Production Center in Fernald, Ohio contain residues from the processing of pitchblende ores. They contain uranium, uranium daughter products, and some heavy metals (primarily lead). Samples of this material will be vitrified, on a laboratory scale, and the off-gas collected, measured, and analyzed for radon and other constituents. Materials, such as lead, which may have been deposited in the off gas system will also be characterized.

TEST DESCRIPTION

The following is a description of the four phases of the test. The systems to be used are shown in Figure 1, Figure 2, and Figure 5.

Phase I During the first phase of the test, the material will be melted in a small inconel "crucible" in a bench scale furnace, and the off-gas will be collected and the volume measured by displacing water from a second vessel. After the crucible has cooled down, the second phase of the test will begin.

Phase II During this phase, the recirculation loop will be valved in, and the recirculation pump will be used to thoroughly mix the total volume of gas in the system. After the gas is well mixed, a sample will be collected in a sample bomb, transfered to the monitoring loop, and analyzed for radon using an Eberline RGM-3 Radon Gas Monitor.

Phase III The third phase involves flushing the system through an activated carbon bed for subsequent radon analysis.

Phase IV During the final phase, the vitrified material will be monitored for radon release using the system shown in Figure 5.

HAZARDOUS MATERIALS AND WASTES

All materials used and wastes produced will be handled in accordance with the Waste Technology Center Chemical and Waste Management Plan and PNL-MA-8. All containers will be labeled appropriately, MSDS's will be available, and personnel will be informed of the hazards present.

SAFETY

Activities associated with the tests will be in accordance with the Waste Technology Center Safety Plan and all applicable SOP's and RWP's. All personnel performing activities are required to understand the safety requirements for the work at hand.

PURPOSE OF THE TESTS

The tests will be conducted to characterize the off-gas produced during the vitrification operation, and to determine the amount of radon emitted during storage of the vitrified product. Analyses will include gas chromatograph/mass spectroscopy, ion coupled plasma/mass spectroscopy, etc.

QUALITY ASSURANCE

Testing is to be conducted as Impact Level II work and will be in accordance with QA Plan WTC-060. Analyses of samples will be obtained through Analytical Request Forms for PNL services or through a Statement of Work for off-site services.

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SC

Figure 1. OFF - GAS COLLECTION SYSTEM

GAS SAMPLE BOMBS

(GS)

TO HOOD

ACTIVATED
CARBON
COLUMN

RECIRCULATION
LOOP

FLOW
METER

INSTRUMENT
AIR

Desiccant

(P)

(P)

COLLECTION
SYSTEM

(T)

(CS)

SLURRIED
K65 MATERIAL
AND
MELTING
CRUCIBLE

(T)

(T)

(T)

(T)

GRADUATED
CYLINDER

COLLECTION VESSEL

FURNACE
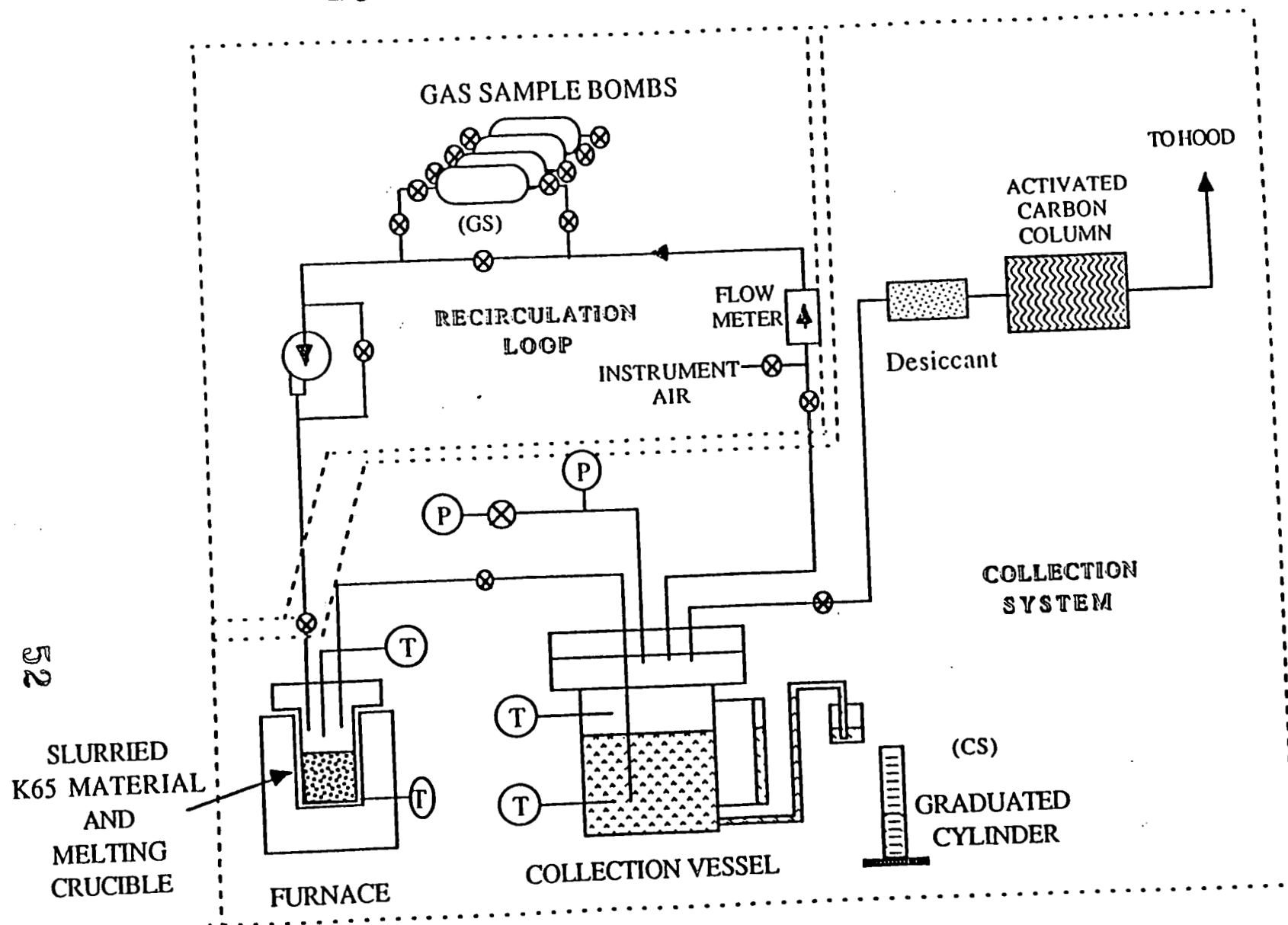
Figure 2. ON SITE RADON MONITORING SYSTEM
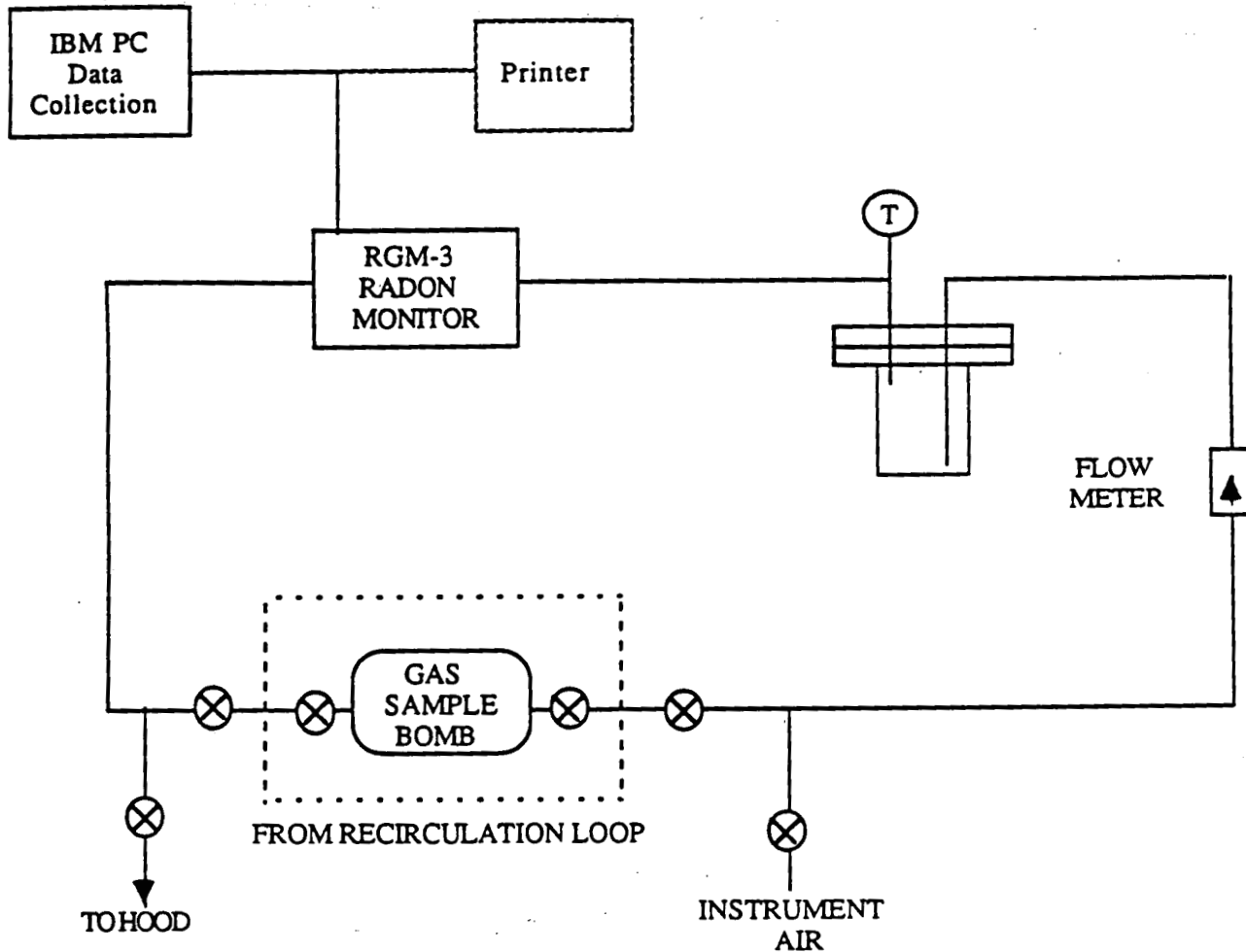
TEST NUMBER _____

PROCEDURE

Exceptions to this procedure may be required to achieve the test objectives due to unanticipated events. Any modifications will be noted either in the following text and/or logged in the operations log which is attached at the back of this document as Data Sheet F. This log should contain any observations that are either unanticipated or which may influence the results of the experiment. It can also contain speculative notes.

Phase I

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35

I. Complete Test System Readiness

The following is a check list of items that should be present and given an operability check prior to beginning the test.

Date   Initials

_____   _____   Furnace

_____   _____   Furnace Temperature Controller

_____   _____   Crucible with sealing flanges and unique label on each flange

_____   _____   Collection Vessel

_____   _____   Collection Vessel and Crucible Seals

_____   _____   Non-evacuated Gas Sample Bombs (75 or 40 ml)

_____   _____   Condensate Sample Bottles (100 ml minimum)

_____   _____   Fresh Activated Charcoal Canister

_____   _____   Glass Former Chemicals

_____   _____   Dilution Canister and Seal

_____   _____   Radon Gas Monitor (and Computer/Printer if required)

_____   _____   Desiccant Cartridge

_____   _____   Flow Meters

_____   _____   Gas Pump

____ ____   Thermocouples and Transducers

____ ____   Collection Vessel Liquid Level measuring device

____ ____   Pressure monitoring devices

____ ____   Miscellaneous supplies: sample labeling tags, Sharpie
              marking pens, work place copy of the procedure, wrenches,
              face shields, leather gloves, high temperature gloves

____ ____   Assemble system as shown in Figure 1 and check for leaks

48 II. Determine the initial amount of gas present in the collection system.
49
50   The purpose of this activity is to allow a calculation of the total
51 number of moles of off-gases generated during the vitrification of the K-65
52 material. To do this, the volume of the system and the temperature must be
53 known at the beginning and end of the test. Pressures will be maintained at
54 nearly atmospheric throughout the test.
55
56   Verify system Readiness
57
58 _____   _____   Set up system as shown in Figure 3. Make certain that the
59   line leaving the collection vessel which drains into the
60   graduated cylinder is completely full of water. Adjust
61   the height of the cylinder so that the water will not
62   syphon in either direction through this line. The water
63   level in the collection vessel should be visible in the
64   sight tube (about 17 liters).
65
66   Determine the volume of the system
67
68   Volume of Crucible Number ____ = _____ (Calculate or by water)
69   Volume of K-65 Material = -_____ (Calculate from weight and density of
70   glass)
71   Total Volume of Piping = _____ (Calculate)
72   Volume of Bomb = _____ (read on Bomb)
73   Volume of Canister = _____ (Calculate or by water)
74   Volume of water in canister at beginning of test = _____ (Calculate)
75   Volume of water in collection vessel at beginning of test = _____
76   (Measured)
77   Total Volume of Collection System = _____ (Sum of Above)
78
79   Volume of Flowmeter = _____ (Calculate)
80   Volume of Pump = _____ (Calculate)
81   Total Volume of Piping = (Calculate)
82   Volume of recirculation loop = _____ (Sum of Above)
83
84
85 III. Obtain and Characterize K-65 Test Sample
86
87   Sample Number (BT-x-mm/dd/yy) = _____   (Note x = 3 or 4; mm/dd/yy
88   is a date format)
89
90   Surface Dose = _____
91
92   Photo Numbers = _____
93
94

Figure 3. VALVE ARRANGEMENT DURING VITRIFICATION

GAS SAMPLE BOMBS

(GS)

RECIRCULATION LOOP

TO HOOD

ACTIVATED CARBON COLUMN

FLOW METER

INSTRUMENT AIR

Desiccant

⊗ VALVE OPEN

● VALVE CLOSED

COLLECTION SYSTEM

P

P

SLURRIED K65 MATERIAL AND MELTING CRUCIBLE

FURNACE

T

T

T

T

COLLECTION VESSEL

(CS)

GRADUATED CYLINDER

2830
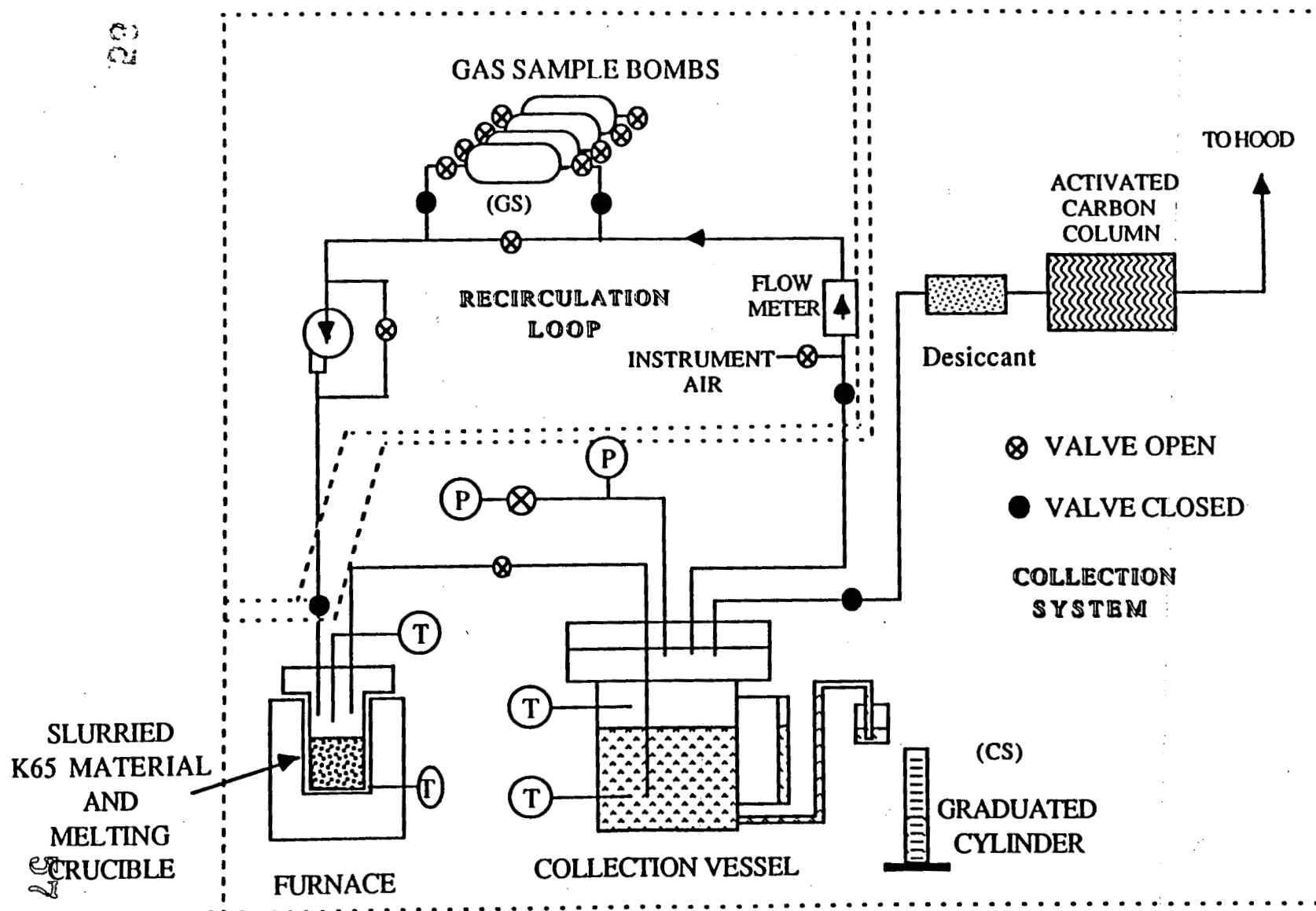
96   IV. Dry and Characterize Test Sample

98   Weight of unfilled sample container _____ units _____.

100   Total weight of sample filled container _____ units _____.

102   Place sample on drying device and allow to dry. Note the beginning time
103   _____ and ending time _____ of the drying process (end drying
104   when sample is greater than 110 °C).

106   Determine weight of sample filled container after drying and record here
107   _____ units _____.

109   Determine net weight _____ units _____.

111 V.   Slurry Sample to Reference Fernald Concentration

113   The reference slurried concentration is XX wt% total solids. Determine
114   the quantity of water needed to slurry the sample as follows:

116   (net weight of dried sample) * ((100/XX)-1) = total weight of water to
117   add

119   _____ / _____ = _____

122 _____ _____   Add this quantity of water to the dried sample and blend
123   to achieve a consistent slurry.

126 VI. Blend in Chemical Additions to Make Glass

128   Chemicals Added (Quantitative and Qualitative Information)

130   Chemical   Quantity

137   Total Weight of Sample, Water, and Chemicals = _____

139 _____ _____   Record weight of empty crucible number ____. _____

141 _____ _____   Place Blend in Crucible Number ____, then seal it by
142   tightening the flange, including a seal, on the canister
143   Close both valves on the crucible.
144 _____ _____   Record weight of crucible and blend. _____ 58

VII. Vitrify K-65 Material and Collect Off-Gases

The goal of this activity is to collect the off-gases during the vitrification of the K-65 material and then to control the dilution, mixing, and sampling of the off-gases so that the original quantity/concentration of radon and other off-gases can be determined.

_____ _____ Verify that valves are as indicated in Figure 3.

_____ _____ Heat filled crucible using the following general schedule: Heat at 200xC for 1 hour, heat from 200 to 1150xC at less than 500xC/hr and greater than 200xC/hr. Hold at 1150xC for a minimum of 2 hours. Record data approximately every 30 minutes on Data Sheet A.

_____ _____ Record time that crucible reached 1150 °C. _____

_____ _____ After a minimum of 2 hours at 1150 °C, shut furnace off. Record time furnace was shut off. _____

_____ _____ Close valve between crucible and canister. Open valves between crucible and pump and between canister and flowmeter as shown in Figure 3a.

_____ _____ Prepare to allow water to syphon back as the crucible cools off.

_____ _____ When crucible approaches 100 °C to 300 °C, record temperature and pressure on Data Sheet A, establish equilibrium conditions between the canister and graduated cylinder, then record the volume of water displaced and the volume of water in the canister.

Volume Displaced = _____
Volume in Collection Vessel = _____

Figure 3a. VALVE ARRANGEMENT AFTER VITRIFICATION DURING COOL DOWN

GAS SAMPLE BOMBS

(GS)

RECIRCULATION LOOP

FLOW METER

INSTRUMENT AIR

TO HOOD

ACTIVATED CARBON COLUMN

Desiccant

⊗ VALVE OPEN

● VALVE CLOSED

COLLECTION SYSTEM

P

P

SLURRIED K65 MATERIAL AND MELTING CRUCIBLE

FURNACE

T

T

T

T

T

COLLECTION VESSEL

(CS)

GRADUATED CYLINDER

2830

60
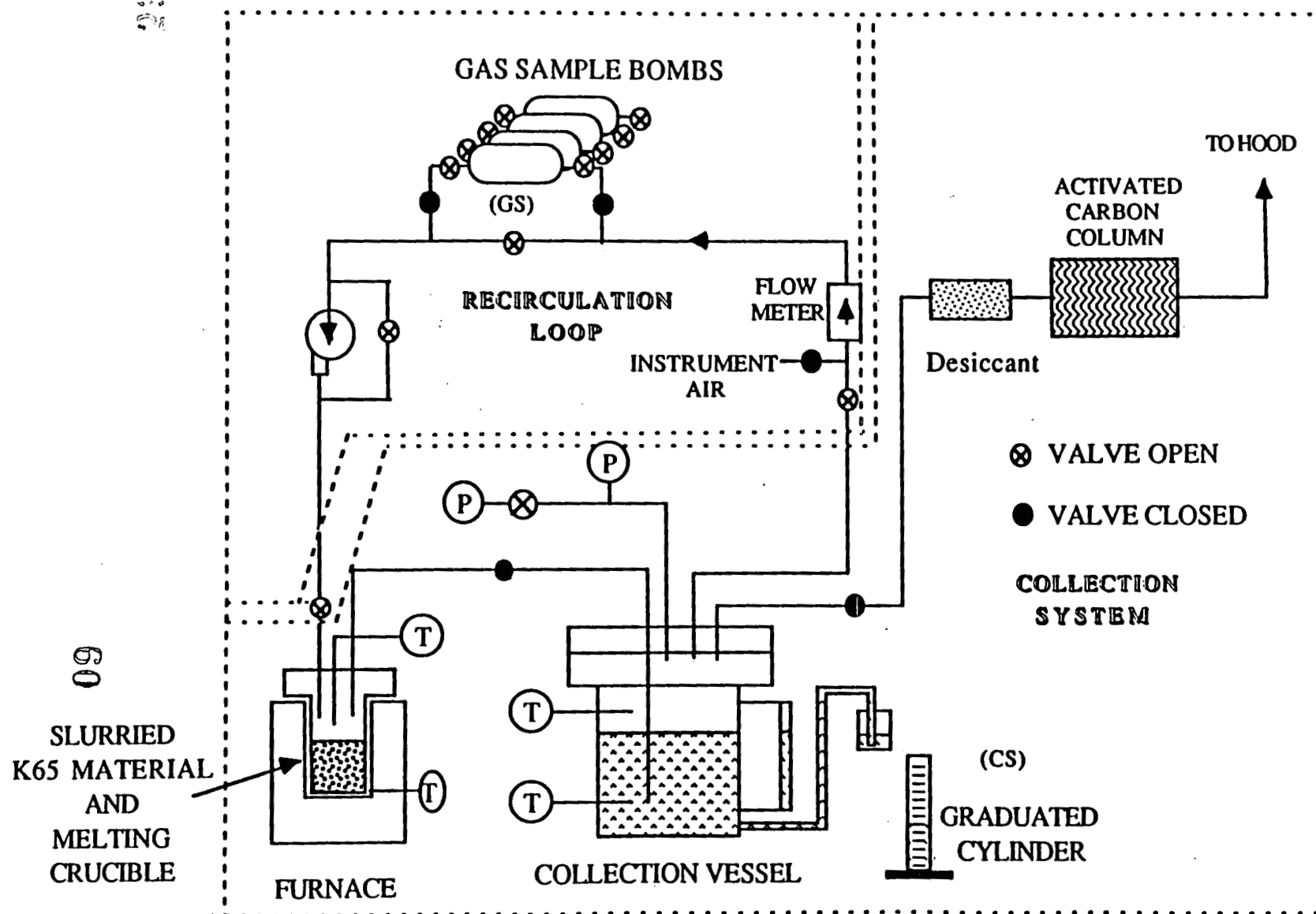
Phase II

_____ _____ Valve in the recirculation loop and turn on recirculating blower/pump as indicated in Figure 3b. Verify recirculation on flow meter. Record flow rate _____ units _____ , system pressure _____ units _____ and time. _____

_____ _____ Ensure 10 system volumes (collection system + recirculation loop), have been pumped.

> 10 volumes = _____
> Divide the 10 volumes by flow rate = _____ = number of minutes to achieve 10 volume changes.

_____ _____ Isolate gas sample bomb by closing the bomb's valves and the isolating valves. Remove from loop and insert into on-site radon monitoring loop as indicated in Figure 2.

_____ _____ Take three gas samples (40 or 75 mL) using the setup shown in Figure 3b and label them as follows: FK65-mm/dd/yy/hh:mm-GSA. Send them for chemical analyses. (Note that mm/dd/yy is a date format and hh:mm is a time format)

_____ _____ Sample #1 tagged and logged into table E

_____ _____ Sample #2 tagged and logged into table E.

_____ _____ Sample #3 tagged and logged into table E.

_____ _____ Turn off recirculation pump in recirculation loop.

Figure 3b. VALVE ARRANGEMENT AFTER VITRIFICATION and COOL DOWN WHILE RECIRCULATING

GAS SAMPLE BOMBS

(GS)

TO HOOD

ACTIVATED
CARBON
COLUMN

RECIRCULATION
LOOP

FLOW
METER

INSTRUMENT
AIR

Desiccant

⊗ VALVE OPEN

● VALVE CLOSED

COLLECTION
SYSTEM

P

P

P

T

T

T

T

(CS)

SLURRIED
K65 MATERIAL
AND
MELTING
CRUCIBLE

FURNACE

COLLECTION VESSEL

GRADUATED
CYLINDER

62
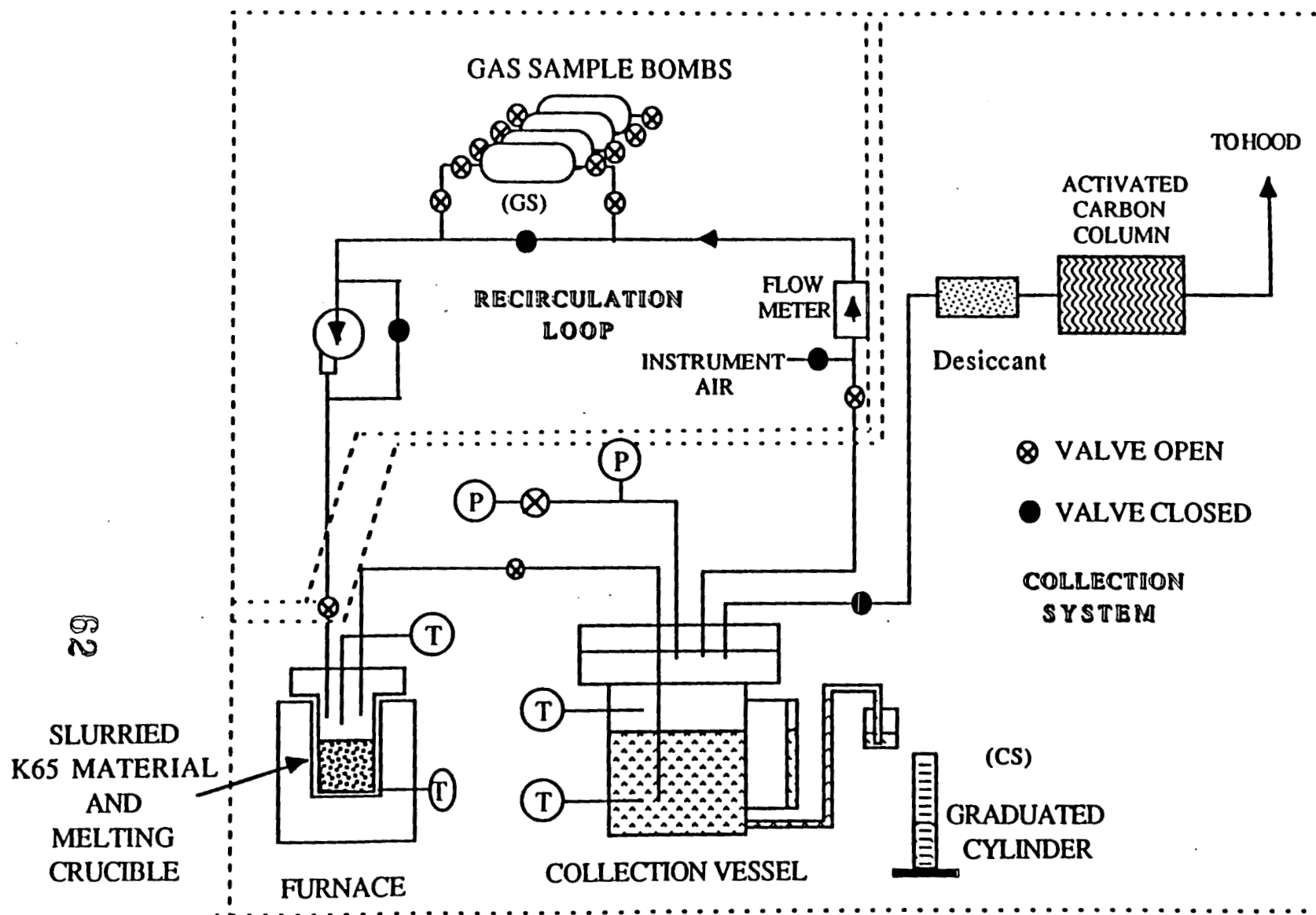
VIII. Determine Volume of Radon Monitoring Loop

The volume of the monitoring loop must be known in order to know how much the gas sample is diluted.

_____ _____ Determine the volume of the monitoring loop.

Volume of dilution canister = _____ (calculate or by water)

Volume of monitor chamber = _____ (given or calc.)

Volume of piping = _____ (calculate)

Volume of Bomb = _____ (read on bomb)

Total volume = _____ (sum of above)

IX. Obtain Radon Gas Concentrations With RGM-3 Monitor

_____ _____ Ensure valves on sample bomb are still closed and open valves to hood and instrument air. Allow monitor to flush (8 minutes). After the flush, the background count will begin (10 minutes), then record the background reading.

_____ _____ Record temperature and radon readings on Data Sheet B approximately every 10 minutes. Verify temperature is below 50 °C. Note that the radon monitor will be damaged if exposed to air hotter than 50 °C.

_____ _____ Close the valves to instrument air and hood and open valves to sample bomb.

_____ _____ Start up radon gas monitor in continuous mode. (An 8 minute pre-sample will be taken, imediately followed by a one hour sample. This one hour sample should be sufficient unless displayed values are erratic.)

Phase III

266
267
268
269   **X.**   **Collect the Radon on a Charcoal Bed and Send for Analysis**
270
271
272   _____   _____   Check condition of the dryer in the charcoal collection
273   system.
274
275   _____   _____   Prepare system valve line up for charcoal bed filtering of
276   gases as indicated in Figure 4.
277
278   _____   _____   Open valves to charcoal collection system and use
279   instrument air to flush collection system and the
280   recirculation loop through the charcoal cartridge. Flush
281   both loops for approximately 15 minutes. Record the
282   temperatures and flow rates on Data Sheet C.
283
284   _____   _____   After flushing the system, valve out the instrument air,
285   label the sample as FK-65-mm/dd/yy-hh:mm-ACS, then place
286   the charcoal cartridge in a sealed package and label the
287   package as FK-65-mm/dd/yy-hh:mm-ACS. Send sample for
288   analysis. (Note that dd/mm/yy is a date format and hh:mm
289   is a time format)
290
291   _____   _____   Flush out crucible with instrument air for
292   (total crucible volume x 10) / flow rate
293
294   (_____ x 10)/ _____ = _____
295
296   close the valves on the crucible, and disconnect it from
297   system.
298
299   _____   _____   Remove flange, vacuum inside spall from the inside,
300   replace high temperature seal with new gortex seal and
301   secure the crucible (bolt flanges and valve crucible
302   closed).
303
304   _____   _____   Record the time the crucible was sealed up. _____
305   Record the crucibles' unique identification number
306   _____.
307

Figure 4. VALVE ARRANGEMENT DURING CHARCOAL FILTRATION
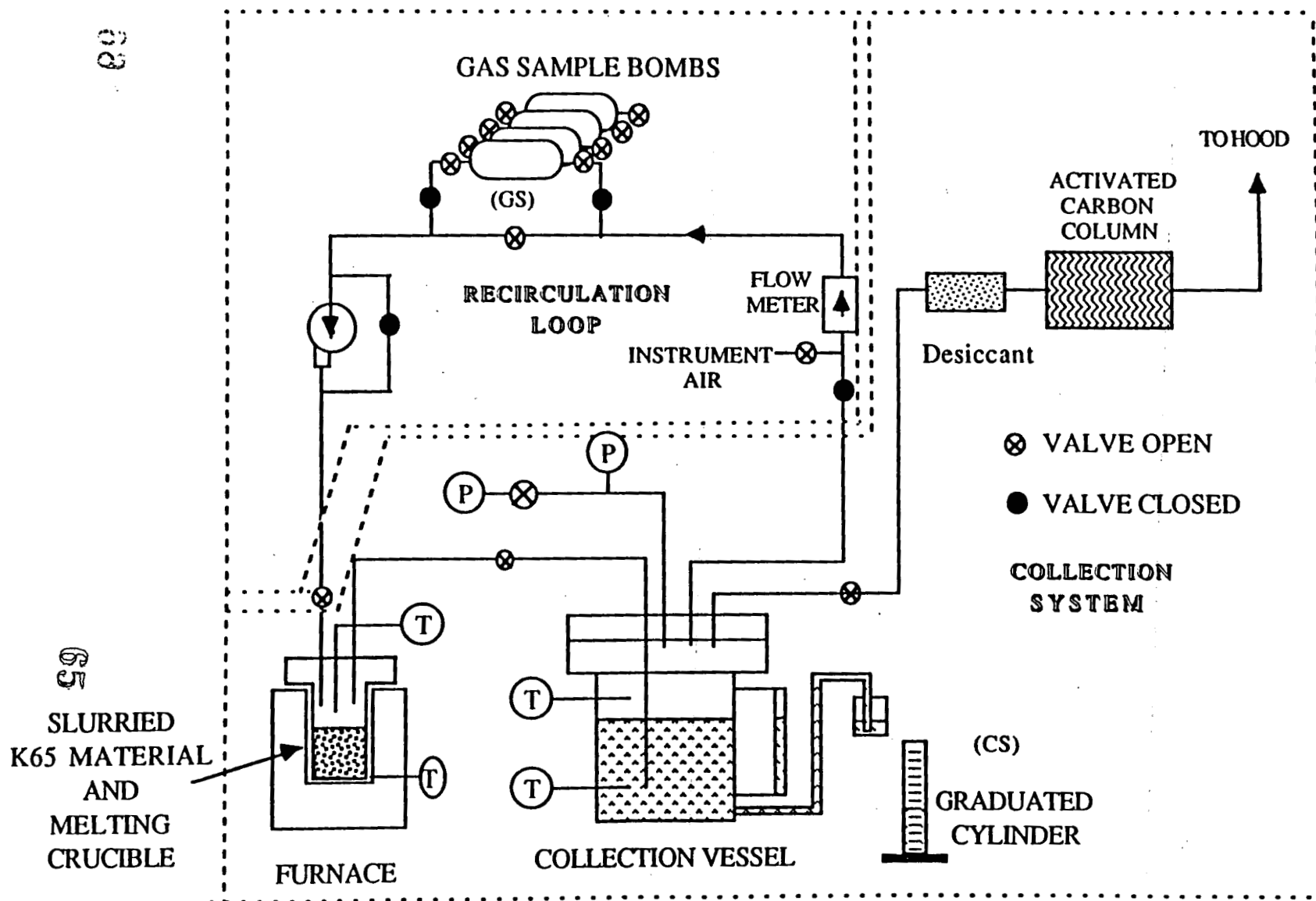
309 XI. **Obtain a Sample of the Condensate and of Materials Plated Out in the**
310 **System**
311
312 ** Note these steps can be completed in parallel with the on site radon
313 monitoring activity.
314
315 _____ _____   Obtain at least a 100 ml sample of the condensate and
316   label it FK-65-mm/dd/yy-hh:mm-CS.
317
318 _____ _____   Send for analyses.
319
320 **NOTE: Do this next step only after all tests have been completed.**
321
322 _____ _____   Dissolve any materials deposited in the pipe between the
323   crucible and the condenser. Obtain a sample and label it
324   FK-65-mm/dd/yy-hh:mm-POS.
325
326
327
328
329   Phase IV
330
331
332 XII. **Post Vitrification Monitoring**
333
334 After all the K-65 material has been vitrified, the following will be used to
335 monitor the filled canisters.
336
337 _____ _____   Set valves in the monitoring system as indicated in
338   Figure 5.
339
340 _____ _____   Ensure valves on crucible number ____ are still closed and
341   open valves to hood and instrument air (Figure 5). Allow
342   monitor to flush (8 minutes). After the flush, the
343   background count will begin (10 minutes), then record the
344   background reading. _____
345
346 _____ _____   Record temperature and radon readings on Data Sheet D
347   approximately every 10 minutes. Verify temperature is
348   below 50 °C. Note that the radon monitor will be damaged
349   if exposed to air hotter than 50 °C.
350
351 _____ _____   Close the valves to instrument air and hood and open
352   valves to crucible as shown in Figure 5a.
353
354 _____ _____   Connect crucible number ____ to the monitoring loop open
355   crucible valves to monitoring loop as in Figure 5a.
356
357

_____   _____   Start up radon gas monitor in continuous mode. (An 8 minute pre-sample will be taken, imediately followed by a one hour sample. This one hour sample should be sufficient unless displayed values are erratic.)

_____   _____   Record the radon readings on Data Sheet D and down load to file or printer and run as long as possible.

Prior to switching to a different crucible, <u>assure that canister is valved closed.</u> Then repeat procedure listed above under Post Vitrification Monitoring.

Just prior to shipping the vitrified sample back to Fernald, determine the non-glass volume of crucible.

_____   _____   Remove flange and fill crucible up with water and determine the volume of water.

_____   _____   Reseal crucible, purge with air, heat crucible at above 100xC for 30 minutes. Then close the canister and make ready for shipment.

Figure 5. RADON MONITORING SYSTEM AFTER VITRIFICATION: FLUSHING AND BACKGROUND MEASURMENT
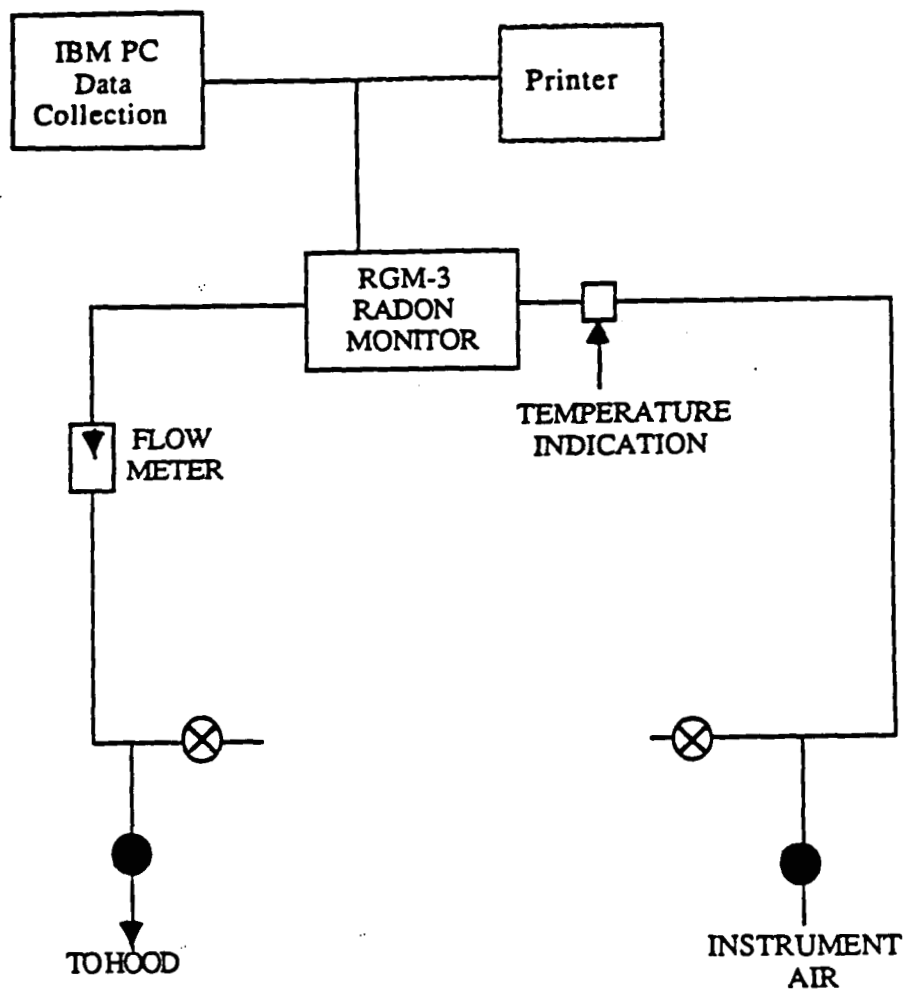
Figure 5a. RADON MONITORING SYSTEM AFTER VITRIFICATION: LONG TERM RADON RELEASE FROM GLASS

CRUCIBLE OF VITRIFIED
K65 MATERIAL

Date	TIME	INI	TEMPERATURE	FLOW RATE	PRESSURE	COMMENTS

DATA SHEET "A" VITRIFICATION OF SAMPLE

Phase I

TEST NUMBER _____

Date	TIME	INI	TEMPERATURE	FLOW RATE	PRESSURE	COMMENTS

DATASHTS.XLS

DATA SHEET "B" ON SITE RADON MEASURING SYSTEM

Phase II

TEST NUMBER _____

Date	TIME	INI	TEMPERATURE	FLOW RATE	RADON CONC.	COMMENTS

DATA SHEET "C" CHARCOAL COLLECTION ARRANGEMENT

Phase III   **TEST NUMBER** _____

Date	TIME	INI	TEMPERATURE	FLOW RATE	COMMENTS

DATASHTS.XLS
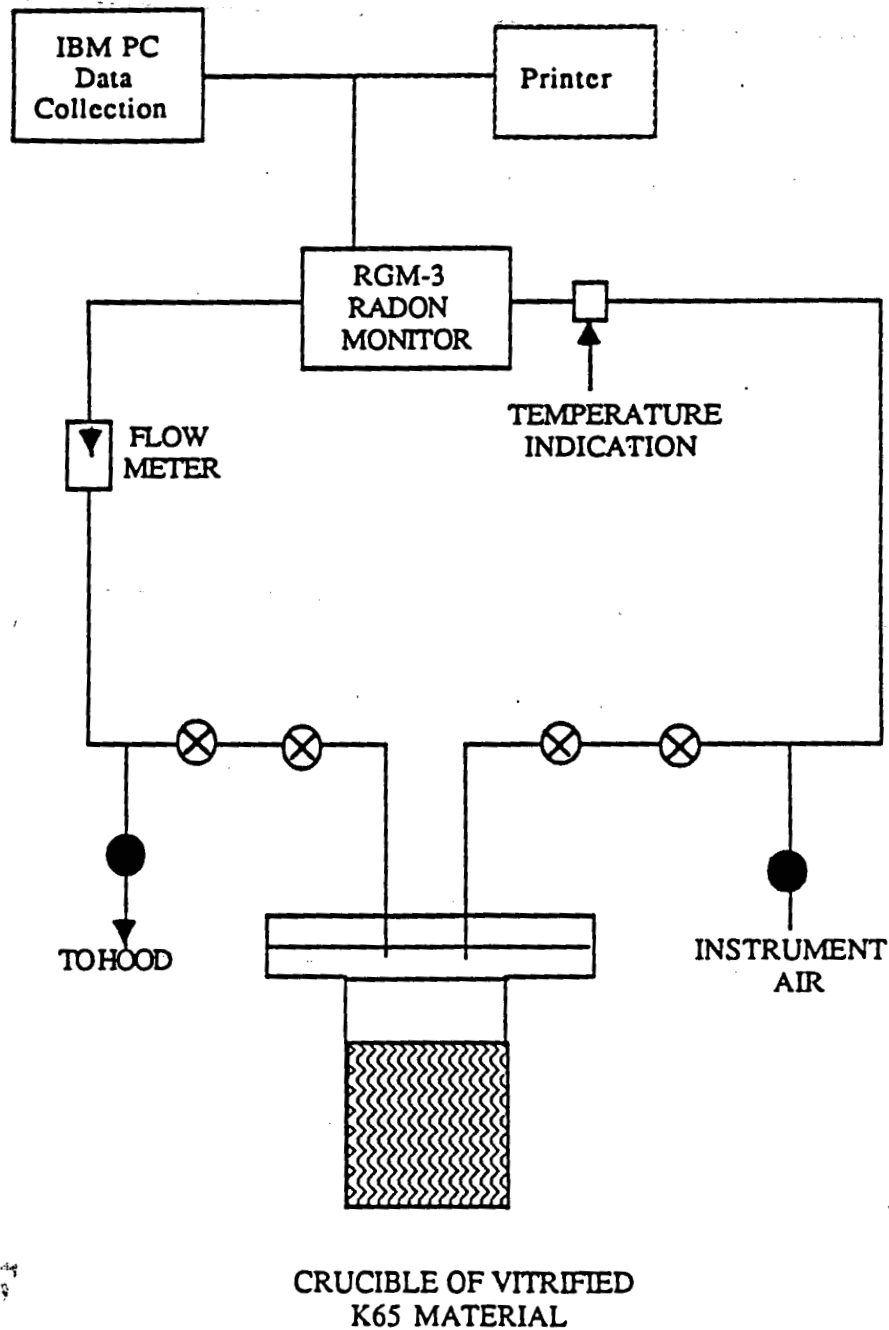
DATA SHEET "D"   RADON CONCENTRATION MONITORING AFTER VITRIFICATION

Phase IV				TEST NUMBER _____		
Date	TIME	INI	TEMPERATURE	RADON CONC.	FLOW RATE	COMMENTS

Date	TIME	INI	DESCRIPTION	SAMPLE ID	COMMENTS

DATA SHEET "E" SAMPLE LOG

TEST NUMBER _____

DATA SHEET "F" OBSERVATIONS

TEST NUMBER _____

DATE	TIME	INI	

APPENDIX B

APPENDIX B

METHODS FOR RADON MONITORING

RADON EMANATION DEPENDENCE ON CONCENTRATION

To verify the effect of radon concentration on the emanation rate, the system shown in Figure 1 will be used. The procedure will be as follows:

- The system will first be leak checked.

- The canister of K-65 residue will be flushed with air for at least five minutes to establish a starting point.

- The pump on the Pylon monitor will be used to circulate the gas around the inner loop (see Figure 1a) while the radon concentration increases.

- After 8 days, the canister of K-65 residue will be valved out, and the radon gas in the sample bomb will be circulated around a dilution loop for at least 25 minutes (see Figure 1b).

- The diluted gas then present in the sample bomb will be further diluted in a second dilution loop (see Figure 1c) where it will also be monitored for radon concentration.

If the concentration measured is significantly less than expected (based on the emanation rate measured during once-through, open system monitoring) the concentration effect will be validated.

OIL ABSORPTION OF RADON

To determine the effectiveness of vacuum pump oil in absorbing radon gas, a filter soaked in the oil will be placed in line with a once-through radon monitoring system (see Figure 2). The data collected with the oiled filter present will be compared to that previously taken over several weeks without the oiled filter present. A concentration difference will verify the oil absorption effect.

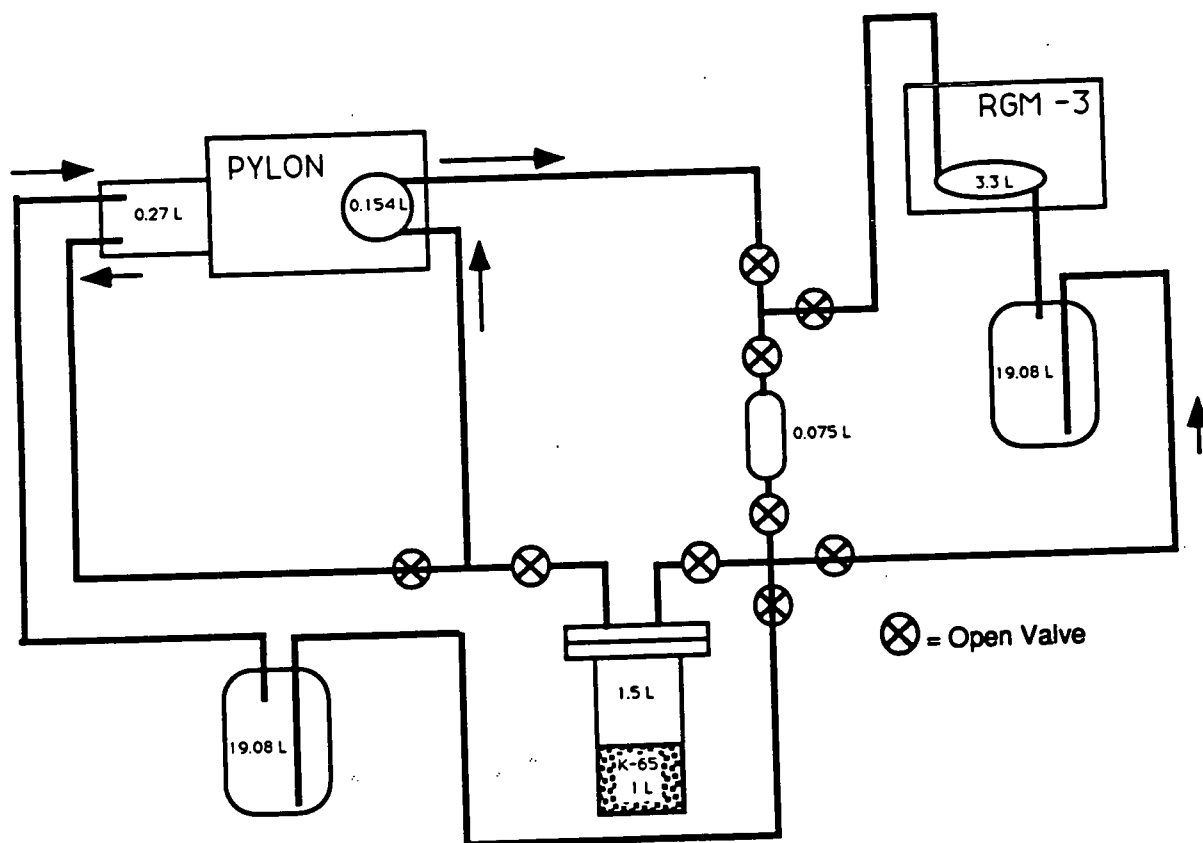## FIGURE 1
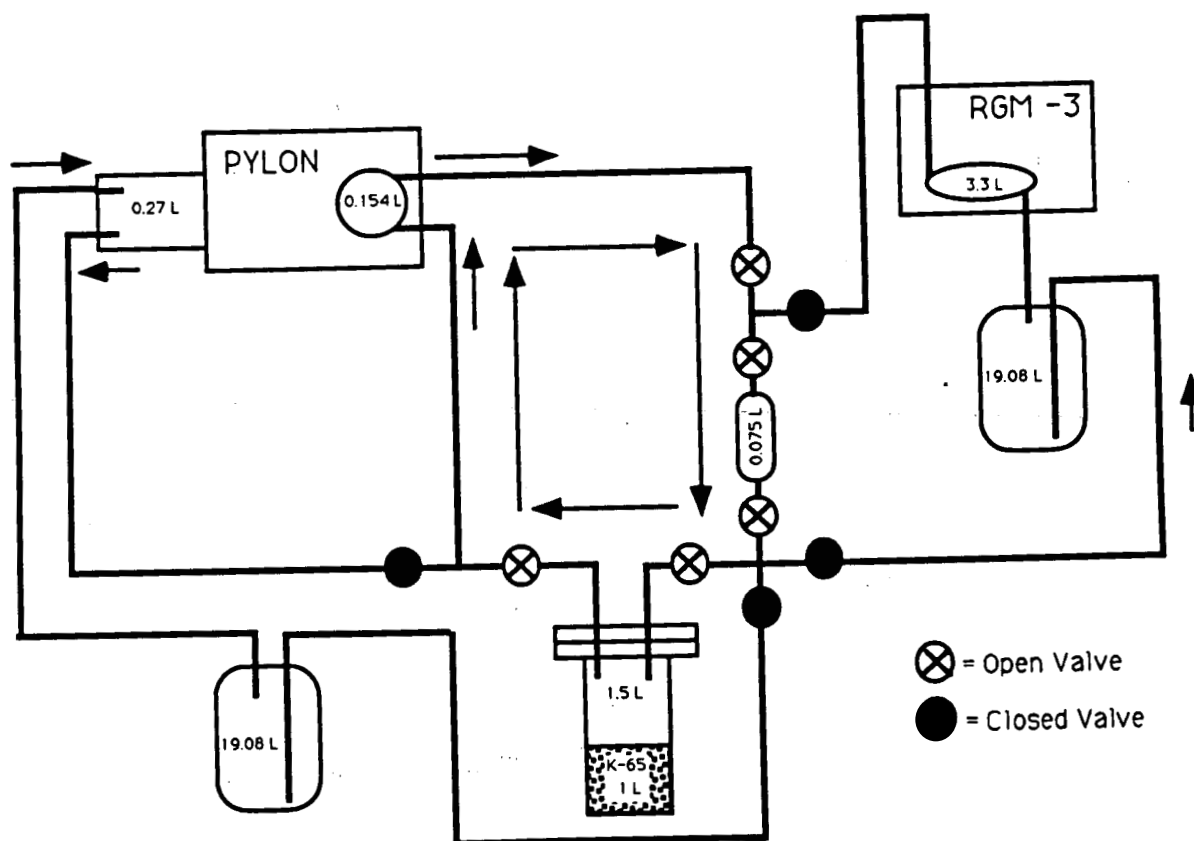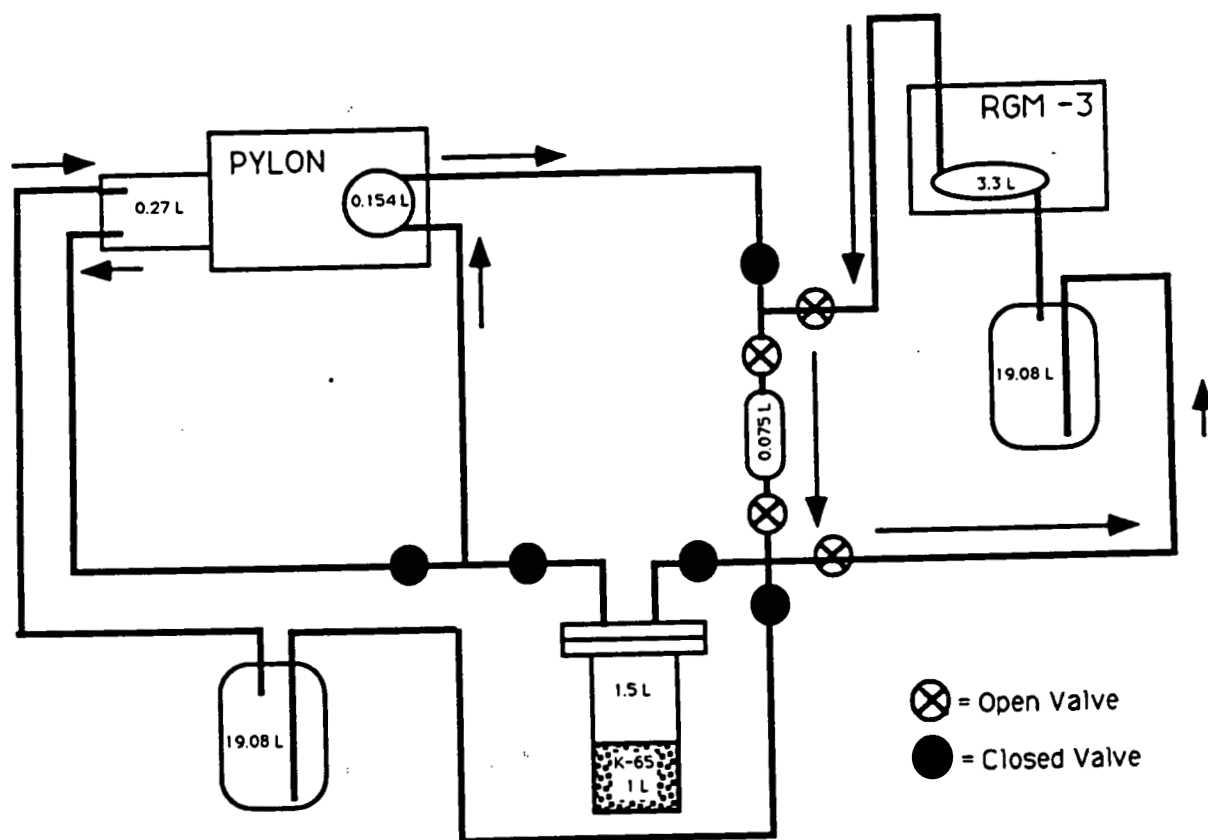### Radon Monitoring System
Nonvitrified K-65 Residue

FIGURE 1a
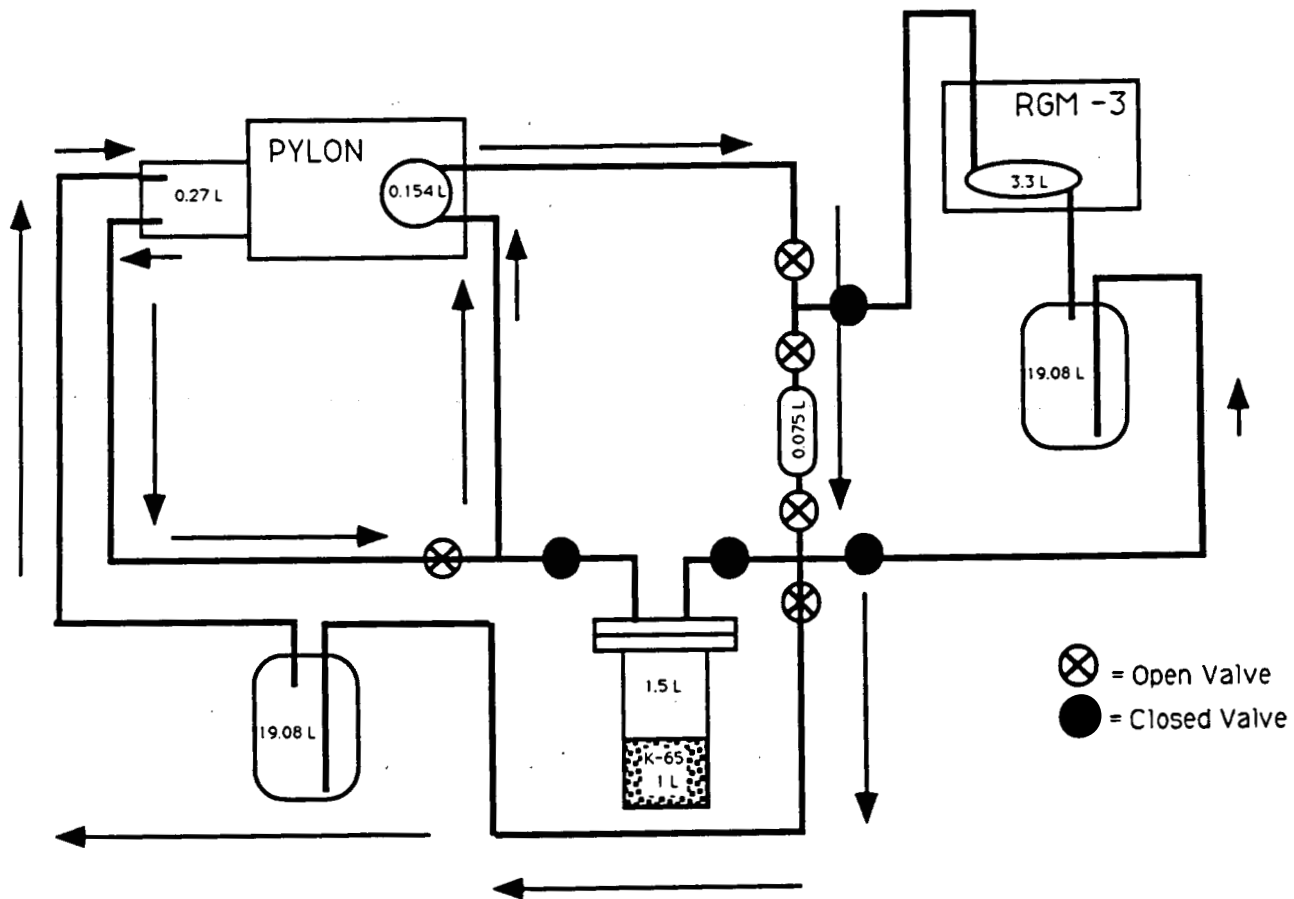## Radon Monitoring System
Nonvitrified K-65 Residue

This configuration allows a homogeneous mixture of radon to build up in a closed loop, and a sample to be captued in a sample bomb

FIGURE 1b
Radon Monitoring System
Nonvitrified K-65 Residue

This configuration allows the sample collected in Figure 1a to be diluted (and possibly monitored)
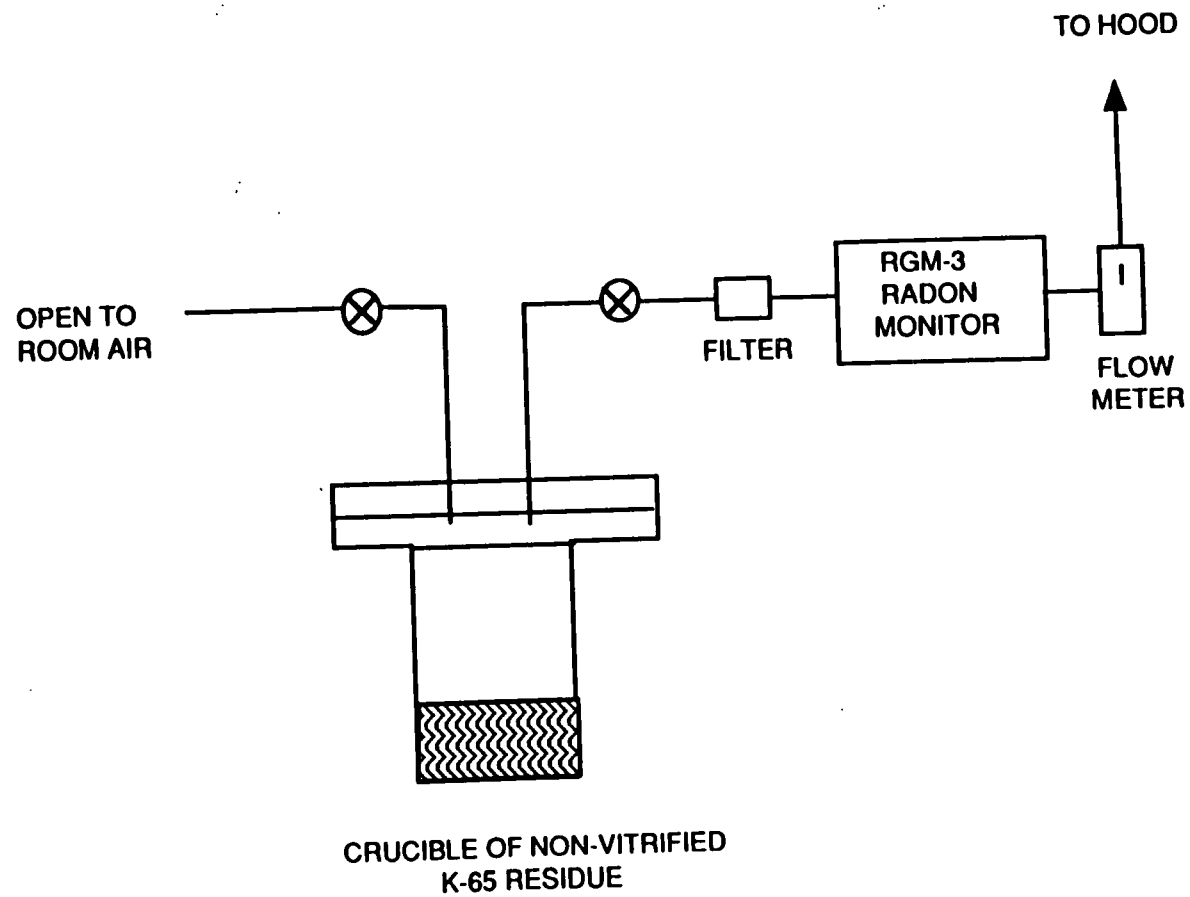
in the loop on the right

FIGURE 1c
Radon Monitoring System
Nonvitrified K-65 Residue

This configuration allows a sample from the dilution loop on the right to be
furthered diluted and monitored in the loop on the left

FIGURE 2.

OIL ADSORPTION RADON MONITORING SYSTEM
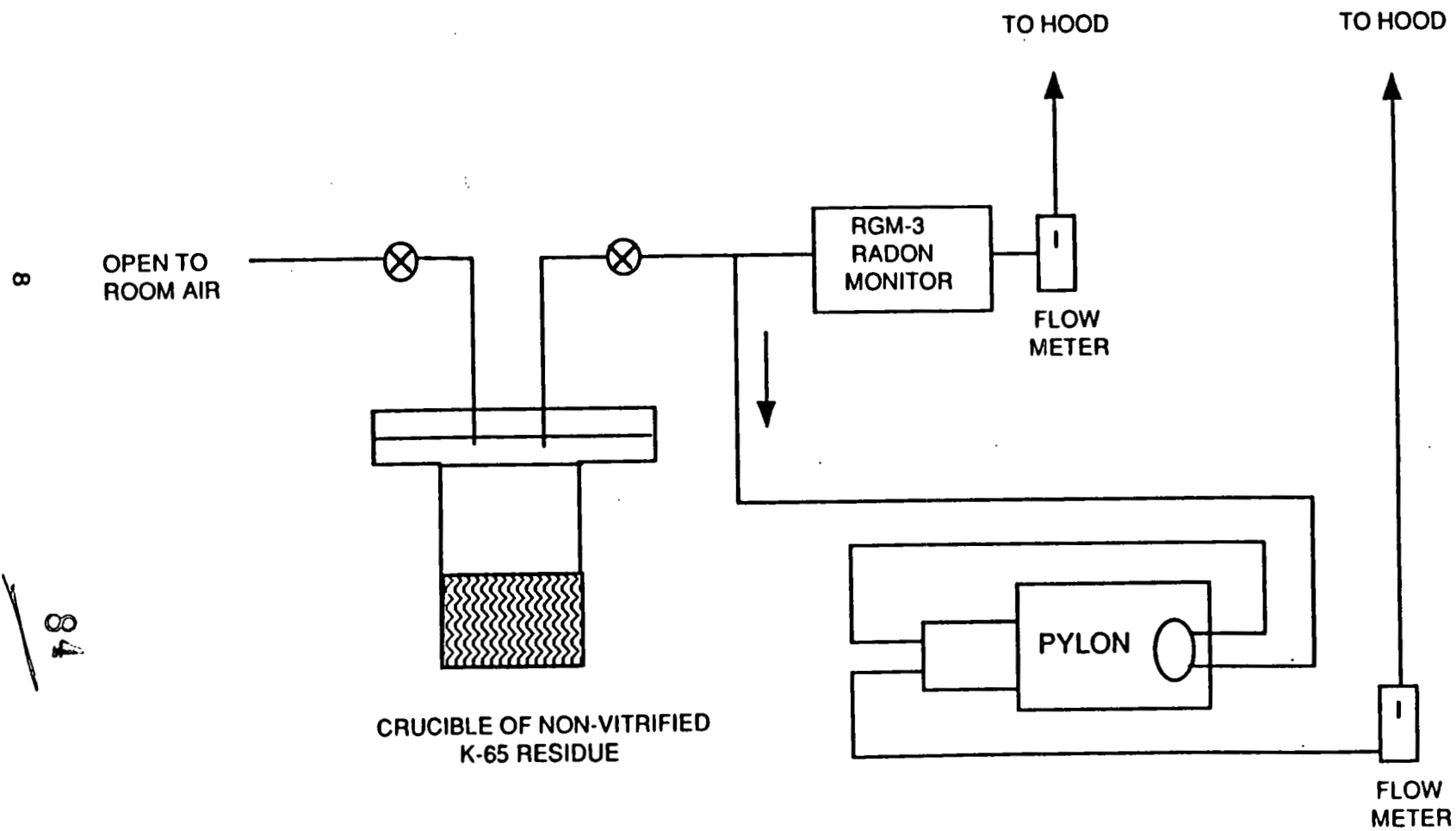

CRUCIBLE OF NON-VITRIFIED
K-65 RESIDUE

THORON EMANATION FROM K-65

To see if thoron is present at a significant concentration, a grab sample of the gas over the K-65 residue will be monitored using the system shown in Figure 3. If the concentration measured decreases during the first 10 minutes of monitoring, then thoron is present.

FIGURE 3

THORON MONITORING SYSTEM

TO HOOD

TO HOOD

OPEN TO
ROOM AIR

RGM-3
RADON
MONITOR

FLOW
METER

PYLON

FLOW
METER

CRUCIBLE OF NON-VITRIFIED
K-65 RESIDUE